

# THE SPIN GLASS TRANSITION

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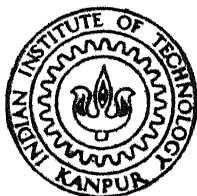
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DEPARTMENT OF PHYSICS

INDIAN INSTITUTE OF TECHNOLOGY KANPUR

SEPTEMBER, 1983

# THE SPIN GLASS TRANSITION

A Thesis Submitted  
In Partial Fulfilment of the Requirements  
for the Degree of  
DOCTOR OF PHILOSOPHY

*By*  
DEBASHISH CHOWDHURY

*to the*  
DEPARTMENT OF PHYSICS  
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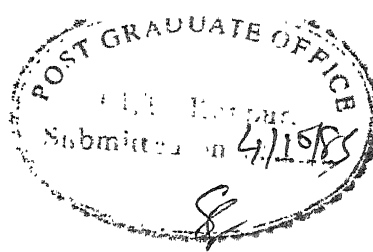
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To

my

parents





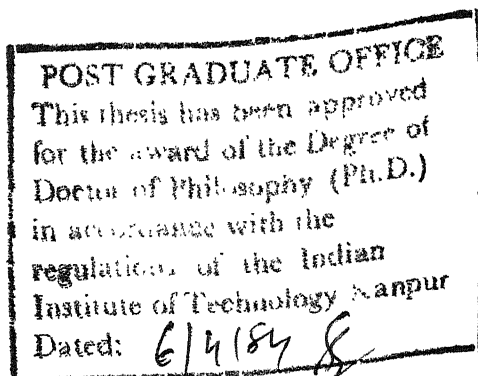
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CERTIFICATE

This is to certify that the work in this thesis entitled, "THE SPIN GLASS TRANSITION" has been carried out by DEBASHISH CHOWDHURY under my supervision. No part of this work has been submitted elsewhere for a degree.

September, 1983.

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## SYNOPSIS

'THE SPIN GLASS TRANSITION'  
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Because of the exchange interaction among the moment bearing magnetic atoms (or ions) in random magnetic alloys, various kinds of magnetic ordering take place depending on the temperature and the concentration,  $c$ , of the magnetic constituent(s). In this thesis we have investigated the nature of the transition from paramagnet-to-spin glass state and the behaviour of various physical properties across the transition.

Recently there have been several attempts to regard a spin glass as the prototype for many more complicated and less understood physical systems. Unfortunately, even the theoretical understanding of the physics of spin glasses is far from complete. There have been two distinct and apparently irreconcilable approaches - the cooperative phase transition model and the model of gradual blocking of superparamagnetic clusters. Both of these have had limited success. All the successes and limitations of these models have been critically reviewed in part A of this thesis.

Part B highlights some further inadequacies of the cooperative phase transition model of Edwards-Anderson type with single relaxation time. Part C proposes an alternative theoretical framework whose predictions are, at least qualitatively, closer to the experimental results. The latter paves the way for an unified approach to the theories of spin glasses by incorporating cooperative effects on the one hand, and features of a gradual freezing process on the other.

Part B begins with Chapter 2 where we investigate the physical conditions under which both Lorentzian and Gaussian local field distributions are good approximations for Ising spin glasses with RKKY exchange interaction. In this chapter we also solve Sherrington-Kirkpatrick equations for two canonical spin glasses numerically and self-consistently so as to find the variation of the order parameters with temperature and external magnetic field.

Mookerjee's theory of magnetoresistance has been applied to two canonical spin glasses in Chapter 3.

Comparing the magnetoresistance thus calculated with the corresponding experimental data we calculate the strength of the s-d exchange interaction,  $J_{sd}$ .

The results of our investigation of relaxation spin dynamics in spin glasses using Master equation approach has been applied in Chapter 4 to calculate the resistivity by Boltzmann transport equation method. The resistivity thus

calculated, using single relaxation time, shows cusp at  $T_g$ , contrary to the smooth behaviour observed experimentally. This is shown to be a consequence of the inadequacy of cooperative phase transition model with single relaxation time.

Chapter 5 is an extension of Chapter 4 to vector spin glasses in the presence of external magnetic field. In this Chapter we improve Mookerjee's theory of magnetoresistance incorporating effects of spin dynamics.

Part C of this thesis begins with Chapter 6 where we develop the percolation model of paramagnet-to-spin glass transition and calculate the transition temperature  $T_g$  as a function of concentration  $c$  taking the effects of finite mean free path and frustration properly into account. In this Chapter we derive Vogel-Fulcher law and explain the variation of  $T_g$  with time and length scales of probes. Besides, we calculate the low-field low-frequency a.c. susceptibility and show that linear response theory holds.

In Chapter 7 the mathematical relation between the percolation model and Wohlfarth's model is established and interpreted physically. Comparing our theoretical results with experimental data we calculate the distribution of blocking temperatures.

The logarithmic relaxation of magnetization is derived analytically in Chapter 8 so as to reveal a deeper connection with  $1/f$  noise in various other physical systems.

Ideas developed in Chapter 8 are extended to calculate the resistivity of spin glasses in Chapter 9.

Generalizing the concept of random walk we calculate the magnetization distribution of spin clusters in Chapter 10 for both classical as well as quantum spins.

In Chapter 11 we prove the existence of a 'hole' in the local field distribution at sufficiently low temperatures. In Chapter 12 we compare our percolation model with some other models heuristically and conclude that "a spin glass is a glass".

LIST OF PUBLICATIONS BASED ON THIS THESIS

- (1) A. Mookerjee and D. Chowdhury,  
Magnetoresistance of AuFe and CuMn: Theory  
and Experiment, J.Phys.F13, 365 (1983).
- (2) A. Mookerjee and D. Chowdhury,  
Spin Glass Transition as a Problem of  
Percolation of Overlapping Spheres,  
J. Phys. F13, 431 (1983).
- (3) D. Chowdhury and A. Mookerjee,  
Relation between Wohlfarth's Model and the  
Percolation Model of Spin Glasses,  
J. Phys. F13, L19 (1983).
- (4) D. Chowdhury and A. Mookerjee,  
Linear Response Theory in a Percolation Model  
of Spin Glasses, Proc. of DAE Nuclear Physics  
and Solid State Physics Symposium,  
Varanasi, India, December 27-31 (1982).
- (5) D. Chowdhury,  
Mean Field Theory of Spin Glasses : A Brief  
Review, Paper Presented at the First Colloquium  
for Young Physicists, organized by Indian  
Physical Society, Calcutta, 16th February, 1983.
- (6) D. Chowdhury and A. Mookerjee,  
Logarithmic Relaxation of Magnetization in  
the Percolation Model of Spin Glass Transition,  
J. Phys. F (1983) (in press).
- (7) D. Chowdhury and A. Mookerjee,  
1/f Noise in Semiconductors and Spin Glasses,  
Solid State Commun. (1983) (in press).
- (8) D. Chowdhury and A. Mookerjee,  
Spin Dynamics in Vector Spin Glasses in  
External Magnetic Field,  
Phys. Lett. A (1983) (in press).
- (9) D. Chowdhury and A. Mookerjee,  
Mean Field Theories of Spin Glasses,  
Submitted to Phys. Reports.
- (10) D. Chowdhury and A. Mookerjee,  
Spin Dynamics and Resistivity of Spin Glasses,  
Submitted for publication.

- (11) A. Mookerjee and D. Chowdhury,  
Magnetoresistance of Spin Glass Alloys-II:  
Submitted for publication.
- (12) D. Chowdhury and A. Mookerjee,  
Cluster Size Distribution and Magnetization  
Relaxation in Spin Glasses,  
Submitted for publication.
- (13) D. Chowdhury and A. Mookerjee,  
Random Walk and Magnetization of Spin  
Clusters in RKKY Spin Glasses,  
Submitted for publication.
- (14) D. Chowdhury and A. Mookerjee,  
Distribution of Blocking Temperatures:  
Percolation Model,  
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- (15) A. Mookerjee and D. Chowdhury,  
Local Field Distribution in Spin Glasses  
Revisited,  
Submitted for publication.
- (16) D. Chowdhury and A. Mookerjee  
Relaxation in Spin Glasses : Effect of  
Nonlinear Terms,  
Submitted for publication.
- (17) D. Chowdhury and A. Mookerjee  
Percolation Model, Sherrington-Kirkpatrick  
Model and Localization-Delocalization  
Model of Spin Glasses,  
Submitted for publication.



## INTRODUCTION

It is usual practice to begin a Ph.D. thesis with a brief review of the existing literature on the subject of investigation followed by a set of emphatic statements that try to convince the reader that the investigation stemmed from a logical analysis, followed a rational course of reasoning and reached a firm conclusion. Besides, no work in theoretical physics can be relevant without comparison with experimental results. Moreover, a detailed critical review (instead of a brief account) of the existing theoretical models often helps in better assessment of their merits and at the same time throws light on their limitations. Therefore, I shall begin with a critical review of the theoretical models, a brief summary of the experimental results together with a comparison of the predictions of the former with the latter in part A.

To achieve maximum logical coherence, details of our original contribution will be presented in two separate parts — B and C; the order being far from chronological. Briefly speaking, part B of this thesis highlights some further inadequacies of the homogeneous mean field theory (with single relaxation time) of cooperative phase transition model of Edwards-Anderson (EA) type for the paramagnet-to-spin glass transition. In part C we propose an alternative

theoretical framework whose predictions are, at least qualitatively, closer to the experimental results. The latter also paves the way for an unified approach to the theories of spin glasses by incorporating cooperative effects on the one hand and features of a gradual freezing process on the other. Some of the details of calculations have been given in Appendices 1 and 2.

PART A

MEAN FIELD THEORIES OF SPIN GLASSES:

A CRITICAL REVIEW

## CHAPTER 1<sup>\*</sup>

### MEAN FIELD THEORIES OF SPIN GLASSES: A CRITICAL REVIEW

#### 1.1 What is a Spin Glass?

The name "spin glass" (hereafter referred SG) was first introduced by Coles [ 1,2] to denote the "entire class of (random) magnetic alloys of moderate dilution in which the magnetic structure no longer resembles that of the pure metal but close enough so that their exchange interactions dominate other energies such as the Kondo effect and other free electron interactions". Such an alloy, "after cooling to a very low temperature in zero applied field has the solute moments "frozen" in local molecular fields. These fields having a distribution of magnitudes and directions such that the net magnetization of any region containing a few tens of solute atoms is zero". So far as the nature of the exchange interaction is concerned, according to Anderson [ 1], "the physical model that has come to be accepted for the substances is that the exchange interaction follows the RKKY theory....the very important property of RKKY which gives SG the unique properties is the oscillation".

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\* A review article based on this Chapter has been submitted for publication in Physics Reports.

This was the picture of SG in 1972. During the last decade many other SG systems have been discovered where the physical origin of the randomness and the nature of exchange interactions are different from the ones considered above. Therefore, Binder [ 3 ] defined SG as "magnetic systems where the interactions between the spins are 'in conflict' with each other, due to some 'quenched (i.e. frozen-in) disorder' in the system". The terms 'quenched disorder' and 'in conflict' will be discussed in detail later. So the essential ingredient<sup>e</sup> of SG is the random nature of the exchange interaction which does not allow the system to exhibit long range order (ferromagnetic or antiferromagnetic) but imposes a new type of order where the spins are thought to be more or less 'frozen' in random directions.

What is the justification for calling these systems 'glass'? In glasses spatial degrees of freedom freeze without any long range order in space. Similarly, the frozen spin degrees of freedom in SG exhibit no long range order. Notice that we have not conclusively stated anything about the dynamical processes of this freezing. Later on we shall see that some particular class of theories draw the analogy between different properties of glasses and SG and argue that SG are also metastable states like glasses. Various aspects of spin glasses have been reviewed in our recent article [ 4 ] and in many other theoretical [ 5-29 ] and experimental [ 30-43 ] review articles.

## 1.2 Spin Glass Systems:

Mydosh [30], and later, Kumar [31] and Ford [35] enlisted and systematically classified the various types of SG. We shall mention some typical examples of each of these classes and then add some more recently discovered ones to the list.

- (a) Noble metal-transition metal alloys: These are called canonical or archetypal SG, typical examples being AuFe, CuMn, AgMn etc. The concentration of the transition metal varies between 0.1 to 10 at %.
- (b) Transition metal-transition metal alloys: The simplest member is PdMn [44]; others include MoMn, RhMn, VFe etc.
- (c) Rare earth SG alloys: Some typical examples are  $\text{La}_{1-x}\text{Gd}_x\text{Al}_2$ ;  $\text{La}_{1-x}\text{Ce}_x\text{Ru}_2$ ;  $\text{La}_{80}\text{Th}_{20}\text{Ce}$ ;  $\text{Ce}_{1-x}\text{Gd}_x\text{Ru}_2$ .
- (d) Transition metal compounds: An extensively studied member of this category is  $(\text{Ti}_{1-x}\text{V}_x)_2\text{O}_3$  [45-50].
- (e) Amorphous Metallic SG: This class [51] includes alloys of the type  $(\text{A}_x\text{B}'_{1-x})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$  where  $\text{A} \equiv (\text{Fe}, \text{Co})$ , and  $\text{B}' \equiv (\text{Mn}, \text{Ni}, \text{Cr}, \text{Mo})$ . Besides amorphous films of GdAl, LaGd etc. also exhibit SG behaviour [52,53].
- (f) Amorphous Semiconducting SG, e.g. amorphous Si, amorphous Ge [54] and  $(\text{Sb}_2\text{S}_3)_x(\text{SbI}_3)_y\text{Fe}_z$  [55].
- (g) Insulating and semiconducting SG: Simplest and widely studied systems are  $\text{Eu}_x\text{Sr}_{1-x}\text{S}$  [56],  $\text{Eu}_{1-x}\text{Gd}_x\text{S}$  [57] and  $\text{Fe}_{1-x}\text{Mg}_x\text{Cl}_2$  [58]. Besides  $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$  and

$\text{Cd}_{1-x}\text{Mn}_x\text{Se}$  [59,60],  $\text{Rb}_2\text{Mn}_{1-x}\text{Cr}_x\text{Cl}_4$  [61],  $\text{CsMnFeF}_6$  [62] and  $\text{PbMnFeF}_7$  [63] also fall in this broad category.

Many insulating amorphous aluminosilicate glasses [64-67] also show SG behaviour.

(h) Intercalated graphite: Intercalating  $\text{FeCl}_3$  into highly ordered graphite gives rise to SG properties of the system [68].

(i)  $\text{PrP}_{0.9}$  is also an induced moment SG [69].

In this thesis we shall be mostly concerned with canonical SG.

### 1.3 Brief Review of Experimental Results:

The experiments performed on SG can be broadly divided into two classes - one of which exhibits a sharp anomaly at a temperature  $T_g$  while the others show a smeared behaviour (a peak or hump) not necessarily around  $T_g$ .

<u>Sharp Anomaly</u>	<u>Smeared Behaviour</u>
Magnetic Susceptibility	Specific Heat
Remanence	Resistivity
Mössbauer	Thermopower
Muon Spin Relaxation	Neutron Scattering
Anomalous Hall Effect	

We shall describe briefly experimental results of only the following properties - Mössbauer effect, magnetization and susceptibility, specific heat, Resistivity

and magnetoresistance, and neutron scattering experiments. Our discussion will be mostly concerned with the experimental results on canonical spin glasses.

### 1.3.1 Mössbauer Experiments:

The Mössbauer experiment measures the local magnetic field at or near a particular site where a radioactive isotope emits or absorbs  $\gamma$ -rays.

Hyperfine splitting, apparently below a sharp temperature, was observed by Violet and Borg [70] and by Window and coworkers [71,72].

Violet and Borg [70] divided the spectra into three classes, depending on their appearance (Figure 1) as follows:

- (I) spectra for which resonance lines are clearly resolved,
- (II) spectra where overlapping of the resonance lines blurs the contrast,
- (III) spectra which appear to be three-peaked.

The areas under the absorption peaks, which is a measure of the line intensities established the random orientations of the frozen spins. Moreover, the total resonance absorption in the unsplit spectra equals that of the split spectra to within  $\pm 10\%$ . This is remarkable because it implies that all of the Fe atoms that exist in the paramagnetic state also exist in the magnetically ordered state thereby establishing the freezing to be a cooperative phenomenon, at least on a time scale of the order



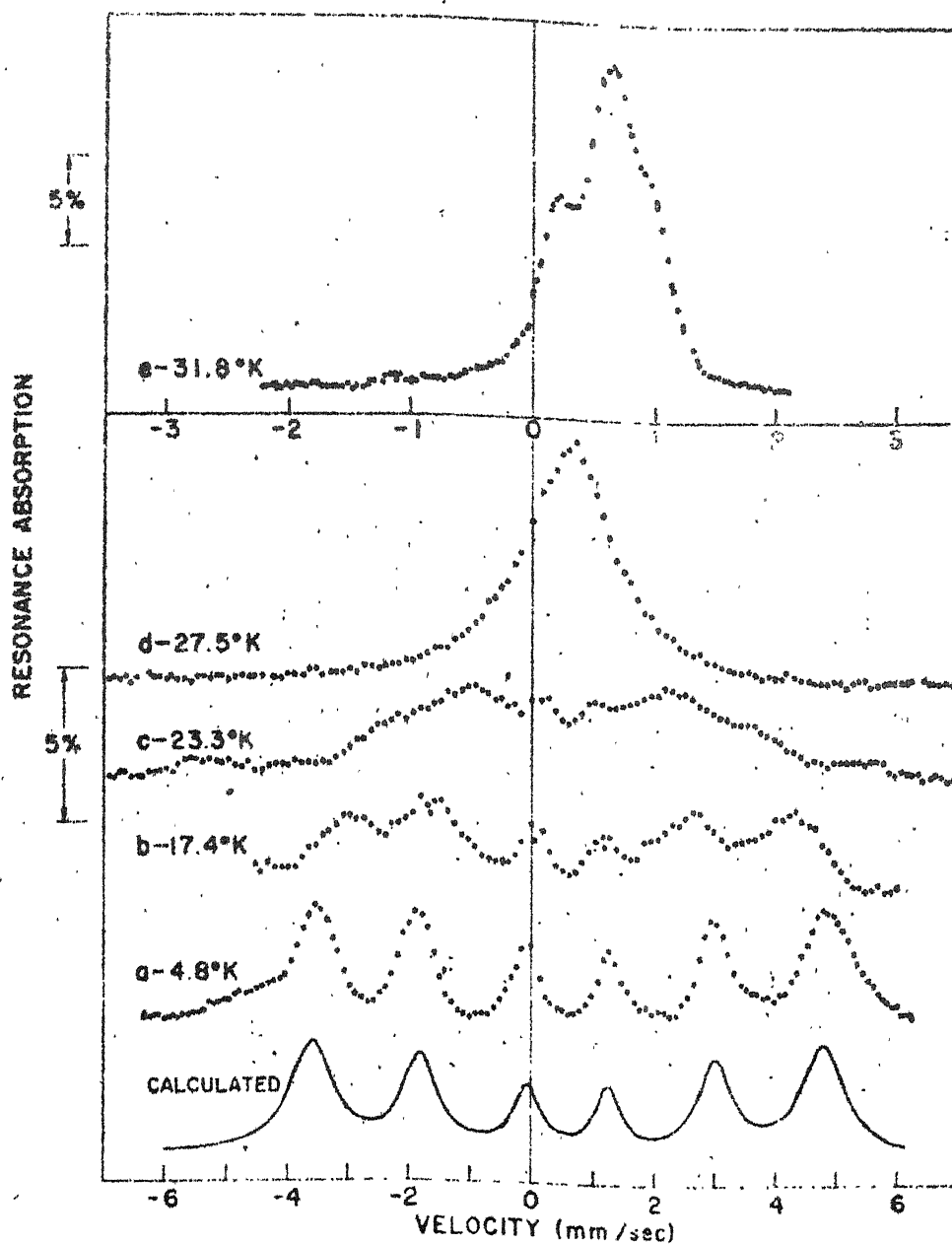


Figure 1 : Resonance Absorption Spectra of Sample AuFe (6.7 at % Concentration) at Various Temperatures (after Ref. [70]).

of  $10^{-8}$  seconds. Last, but not the least, important observation was that there is no detectable unsplit paramagnetic component superposed on the magnetic hyperfine spectra. This implies that below  $T_g$ , the fraction of 'loose' spins having a null hyperfine field is less than 1%.

### 1.3.2 Magnetization and Susceptibility Measurements

There are three aspects of magnetization  $M(H, t, T)$  to be studied -

- i) variation of  $M$  with external magnetic field  $H$  and hysteresis,
- ii) variation of  $M$  with time  $t$  which reveals irreversible and nonequilibrium processes, i.e., magnetization relaxation,
- iii) variation of  $M$  with temperature.

We know that magnetization and external magnetic field are related by response functions called magnetic susceptibility:

$$M = \chi_0 H + \chi_1 H^2 + \dots \quad (1.1)$$

where  $\chi_0$  is the linear susceptibility (or zero field susceptibility) and  $\chi_1, \chi_2$  etc. are nonlinear susceptibilities. Now onwards susceptibility will mean linear susceptibility, if not otherwise stated. But since zero field susceptibility is only an ideal concept not realizable in practice, one measures very low field susceptibility instead.

Cannella and Mydosh [ 73] first made successful measurements at low field (5G) and low frequency (155 Hz) and got the following interesting results:

- (a)  $\chi(T)$  exhibits a sharp cusp apparently at a sharp temperature  $T_g$  as shown in Figure 2,
- (b) The cusp is very sensitively field dependent (Figure 3) and gets flattened even at low fields, eg. 50G. They concluded that the failure of all previous measurements, who observed only broad maximum instead of sharp cusp, was due to the use of higher fields which was unavoidable for limitations of accuracy,
- (c) The maximum of a.c. susceptibility always occurred at a temperature lower than the temperature at which Mössbauer splitting occurs.

Later experiments [ 74,75] showed that  $T_g$  shifts with the frequency of the a.c. measurement (Figure.4). At this point one should note that the frequency of the a.c. measurement can be varied to probe the dynamics of the freezing process over varying time scales.

The significance of using low field is clear. But what is the use of the a.c. technique instead of d.c. measurement? The answer to this question is very closely related to the variation of magnetization with time. Guy [ 76] performed accurate measurement of d.c. magnetization in a field of less than twenty gauss. He observed that

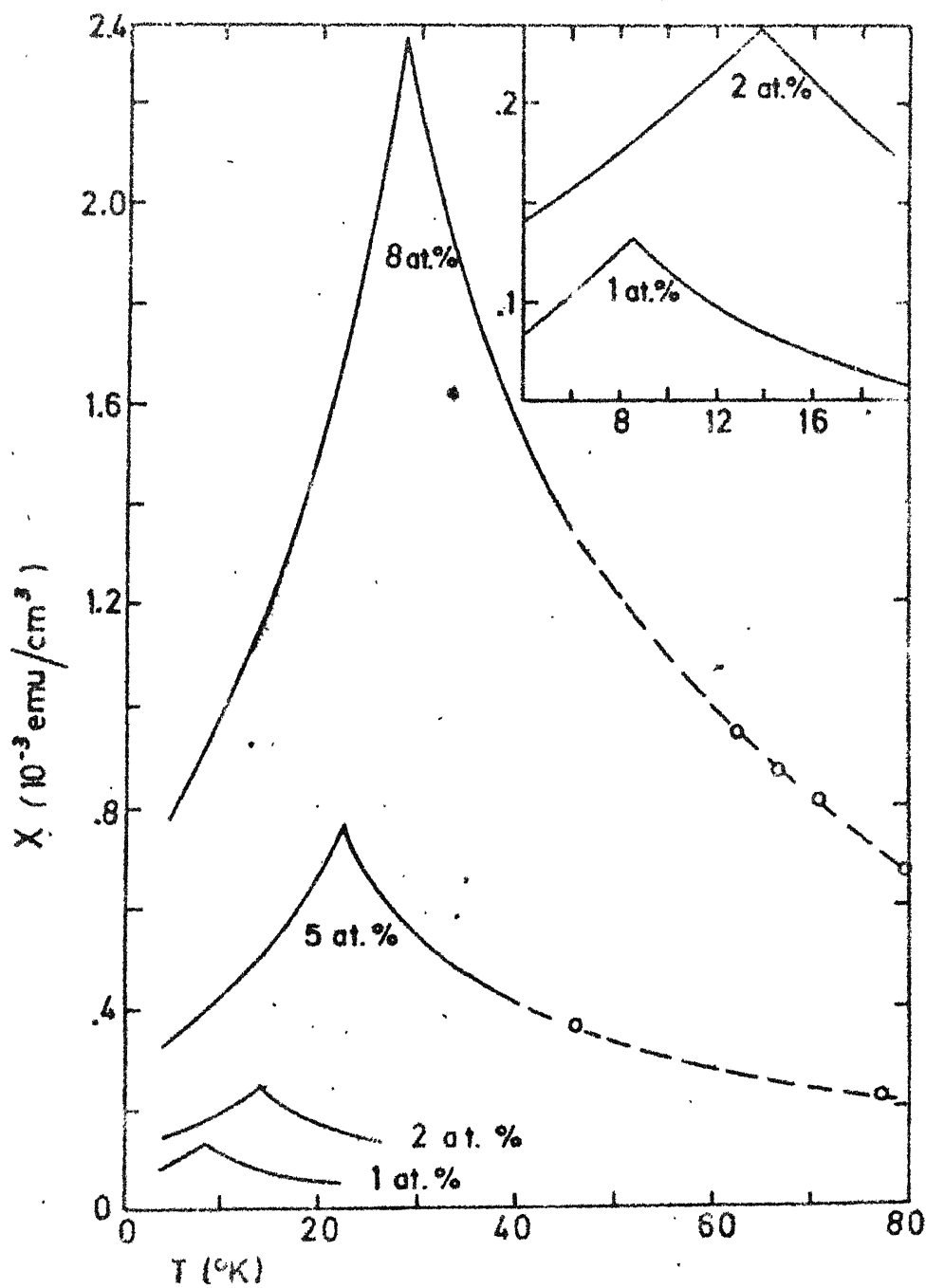


Figure 2 : Low Field Low Frequency a.c. Susceptibility  $\chi(T)$  for  $1 \leq c \leq 8$  at %. (After Ref. [73]).

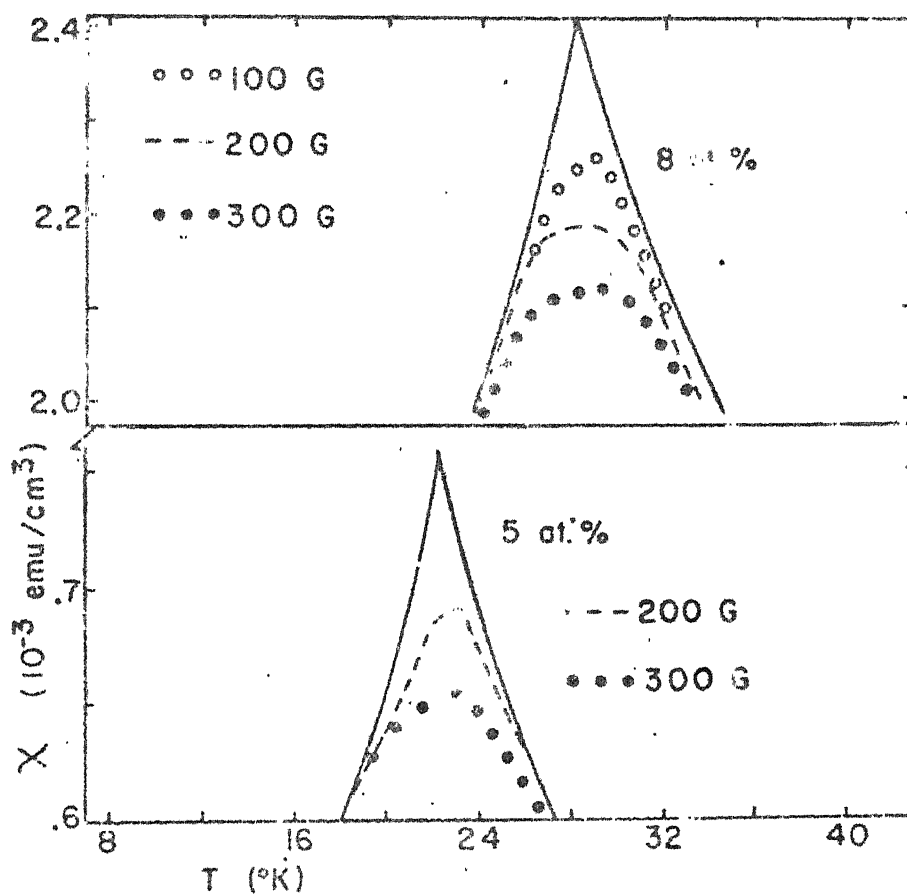


Figure 3 : Susceptibility Data for Samples with  $c = 5$  and  $8 \text{ at. \%}$ , showing the Curves for Zero Field, and for Various Applied Fields (after Ref. [73]).

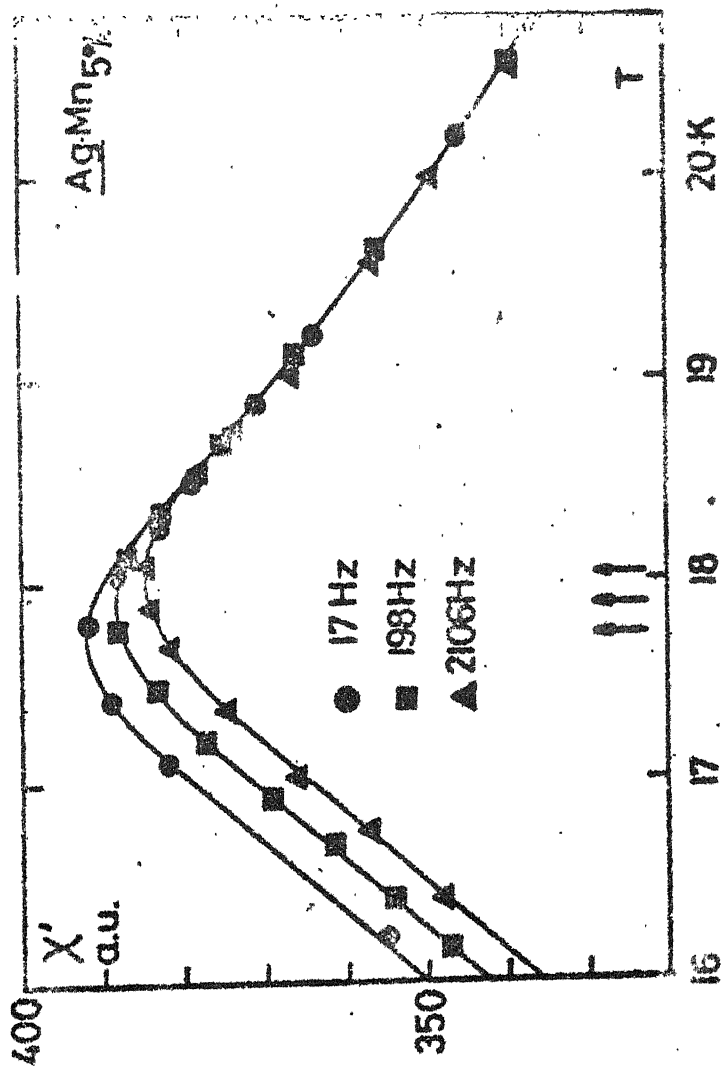


Figure 4 : A.C. Susceptibility of AgMn (5 at %) in the vicinity of  $T_g$  (after Ref. [75]).

- (a) for systems cooled in zero external field magnetization attains a value  $M_0(T)$  within a very short time after the external field is switched on. However, even if the field and temperature are kept constant the magnetization increases slowly with time so that

$$M(t, T) = M_0(T) + \Delta m(t, T) \quad (1.2)$$

On the other hand, if the external field is turned off after a time  $t$ , an IRM is left whose initial value is apparently  $\Delta m(t, T)$  and which decays slowly in nearly the same way as  $\Delta m(t, T)$  develops with the field applied (see Figure.5).

Guy observed that

$$\Delta m(t, T) = S(T) \ln t \quad (1.2a)$$

He distinguished two quantities:

- (i) ideal nonequilibrium magnetization

$$M(H, T, t=0) = M_0(H, T)$$

- (ii) equilibrium magnetization

$$\lim_{t \rightarrow \infty} M(H, T, t) = M_0(H, T) + \lim_{t \rightarrow \infty} \Delta m(t, T)$$

- (b) equilibrium magnetization can be alternatively achieved by cooling in an external magnetic field,
- (c) in the a.c.measurement one measures only the reversible part of the magnetization, and hence the difference between a.c. and d.c. susceptibility.

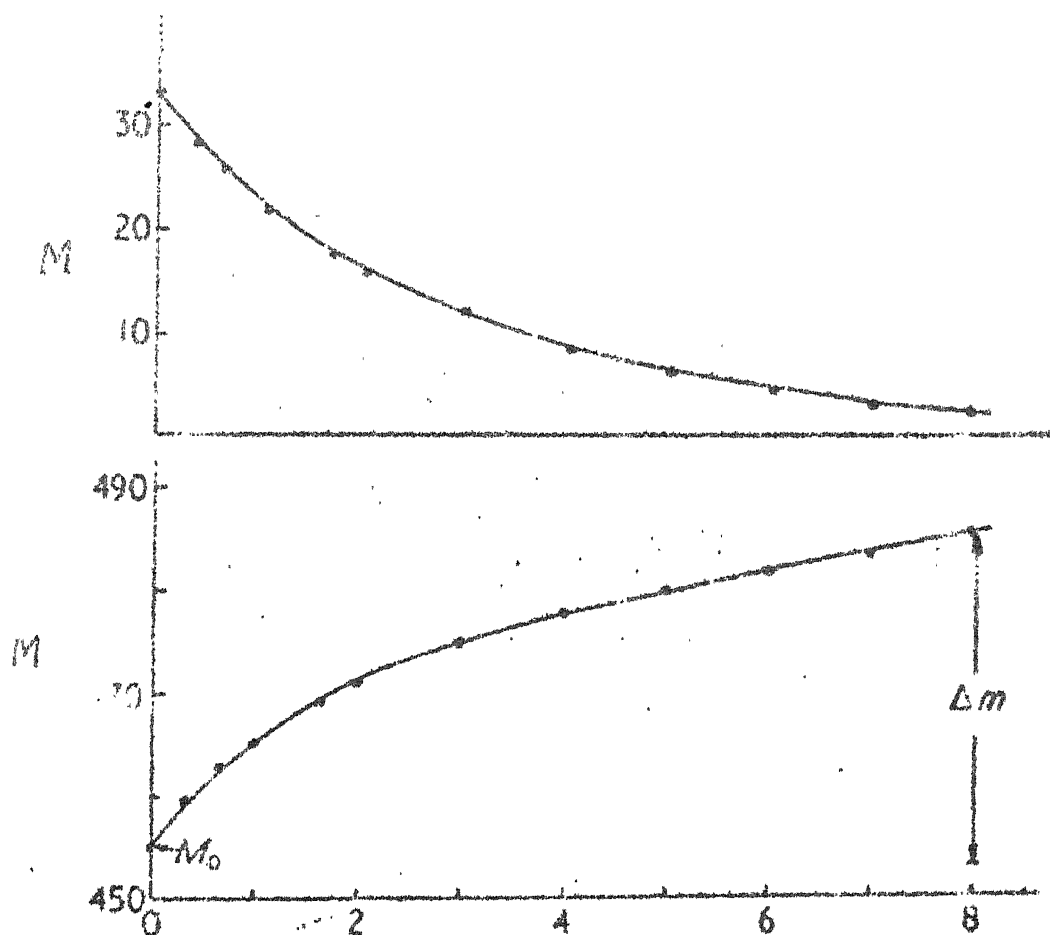


Figure 5 : The Time-dependent Behaviour of Magnetization at 4.2 K. The Lower Curve shows the Development of Magnetization with Time in an Applied Field of 20G. The Upper Curve shows the decay of the IRM obtained after switching off the field of 20G at  $t = 8$  min. (after Ref. [76] ).



The hysteresis curve differs from that of a ferromagnet because the remanent magnetization for a SG is time-dependent and decays to the equilibrium value ( $M(t = \infty) = 0$  for  $H=0$ ) very slowly for all  $T < T_g$ . The other interesting feature of the hysteresis curve is that it is laterally displaced from the origin as shown in Figure 6.

### 1.3.3 Specific Heat Measurements:

First of all, we should recognise that it is extremely difficult to separate out the electronic contribution to specific heat from that of magnetic part because both vary as proportional to temperature  $T$ , although separation of the phonon part is not very difficult as it varies as  $T^3$ . It is generally assumed that magnetic contribution to specific heat = (specific heat of the system containing magnetic impurity) - (specific heat of the system, under exactly same conditions, where magnetic impurities have been substituted by nonmagnetic impurities of atomic number very close to that of the magnetic impurity). It was observed [77]

- (i) that magnetic contribution to specific heat  $C_m$ , exhibits only a broad hump whose maximum occurs at a temperature higher than that of susceptibility, (Figure 7),
- (ii) that a plot of  $C_m/T$  also reveals no anomaly for most of the systems,

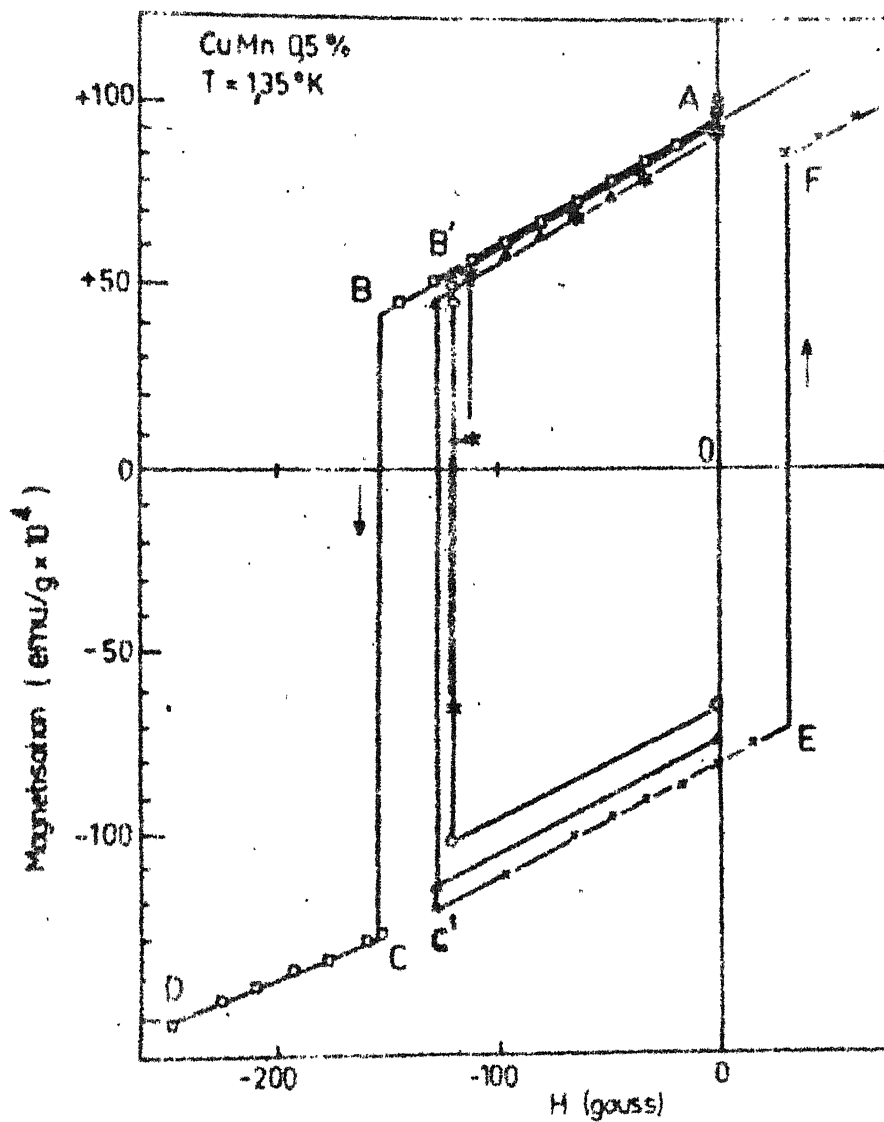


Figure 6 : Hysteresis for field-cooled CuMn with a saturated remanent magnetization. Different symbols are used for different runs (after ref. [33]).

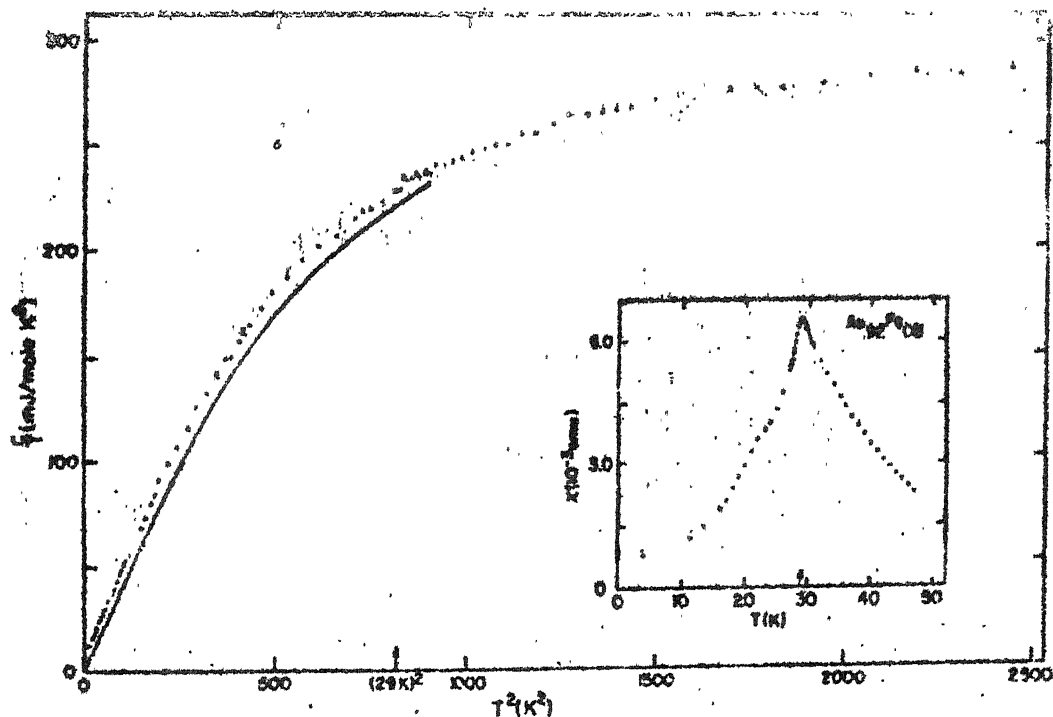


Figure 7 : Specific heat of  $\text{Au}_{0.92}\text{Fe}_{0.08}$  in the temperature region 3-50K. The solid curve is the calculated nonmagnetic contribution to the specific heat of the alloy between 0 and 30K. The insert shows the susceptibility results of the sample (after [77]).

- (iii) that only 30% of the total entropy is recovered upto  $T_g$ , the remaining 70% develops above  $T_g$ ,
- (iv) that the specific heat is linear in  $T$  at very low temperatures.

Very recently Thomson and Thompson [ 78,79] have presented evidence for  $T^{3/2}$  behaviour of low temperature specific heat of SG, exactly as in case of resistivity, to be discussed later. This type of  $T^{3/2}$  behaviour was already suspected by Coles [ 2 ] in 1972.

#### 1.3.4 Resistivity and Magnetoresistance:

Ford and Mydosh [ 80] measured the magnetic or spin resistivity  $\rho(T)$  at temperature  $T$ ,

$$\rho(T) = \rho_{\text{alloy}}(T) - \rho_{\text{pure host}}(T)$$

as shown in Figure 8. However, such a method becomes inaccurate if deviation from Matthiessen's rule becomes appreciably large. Their observations can be summarized as follows:

- (a) residual resistivity  $\rho_0 \propto c$  for lower concentration alloys,
- (b) for  $c \geq 5$  at % variation of  $\rho_0$  with  $c$  becomes slower than linear and at about 12 at %  $\rho_0$  reaches a maximum. On further increase of  $c$ ,  $\rho_0$  gradually levels off at  $c \geq 25$  at % Fe.

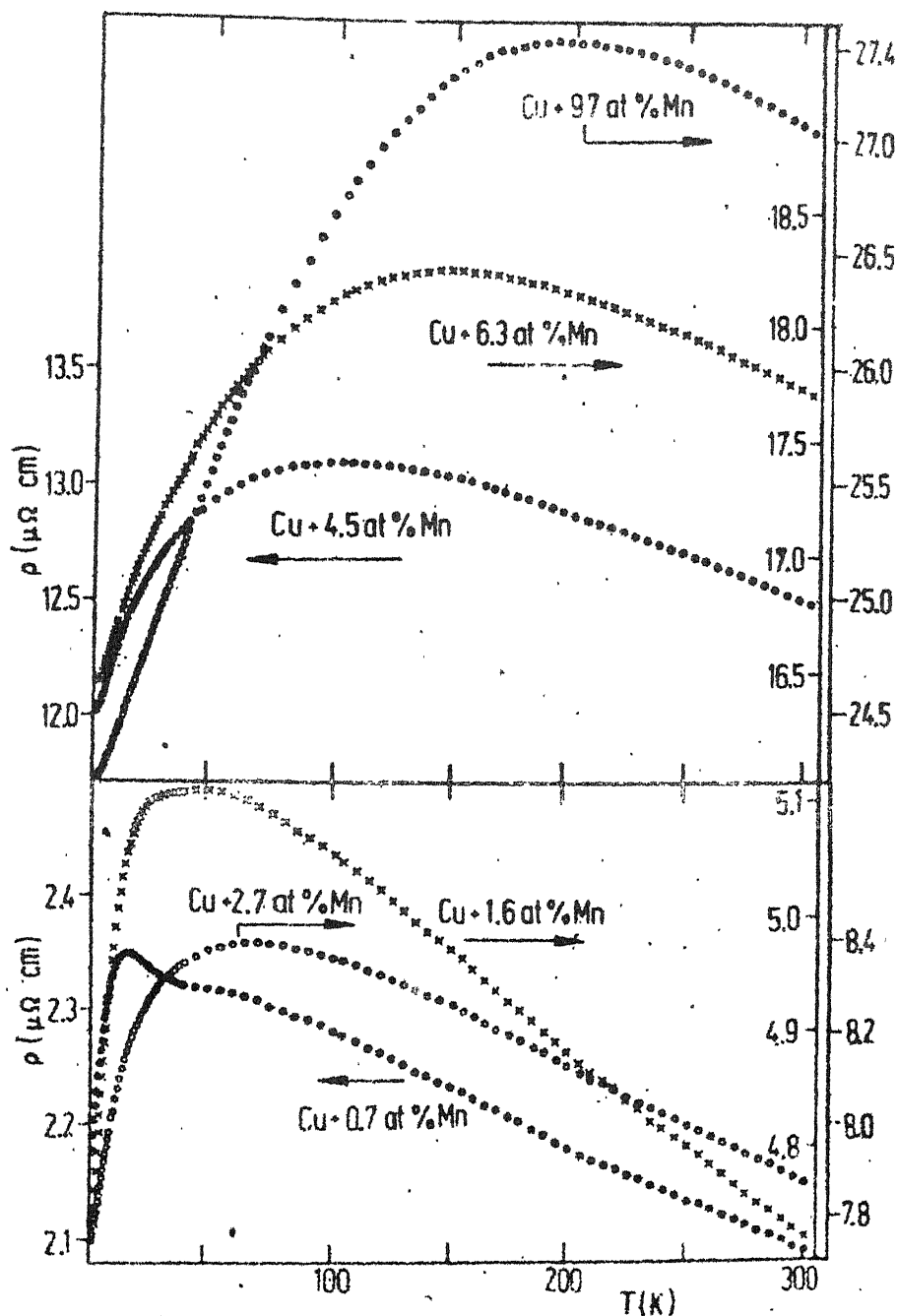


Figure 8 : Over-all temperature variation of  $\rho$  ( $\mu\Omega$  cm) for CuMn alloys with concentrations between 0.7- and 9.7- at % Mn. Note the changes in scale as the concentration increases (after ref. [80]).

- (c) at low temperature  $\rho(T)$  rises steeply, initially faster than  $T$ , and then practically linear with  $T$  in the region of  $T_g$ . With further increase of  $T$ ,  $\rho$  reaches a very broad maximum at a temperature  $T_{\max} \gg T_g$  and then gradually falls.
- (d) upto the lowest temperature of 0.5K best fit with experimental data was obtained with

$$\rho(T) = \rho_0 + A T^{3/2} \quad (1.3)$$

- (e) the range of the  $T^{3/2}$  fit increases with increasing concentration
- (f) the coefficient  $A$  varies with  $c$  as shown in Figure 9
- (g)  $T_{\max}$ , the temperature corresponding to the maximum in  $\rho$ , increases less rapidly than  $c$ , roughly between  $c^{0.4}$  and  $c^{0.6}$ . The qualitative behaviour of  $T_{\max}$  versus  $c$  is similar to that of  $T_g$  versus  $c$ .

Senoussi [81] observed that for zero field-cooled.

AuFe SG the transverse and longitudinal magnetoresistance are the same. He showed that Transverse Magnetoresistance (TMR)/ $\rho$

$$= \frac{\Delta\rho}{\rho} = -\alpha M^2 = -\alpha X^2 B^2 \quad (1.4)$$

where  $\alpha$  is a constant. Senoussi used this expression and Nigam and Majumdar's [82] experimental values of TMR to calculate  $X$ . These calculated values of  $X$  agreed quite well with those measured directly.

The most detailed study of TMR in canonical SG has been made by Nigam and Majumdar [83,84]. They observed

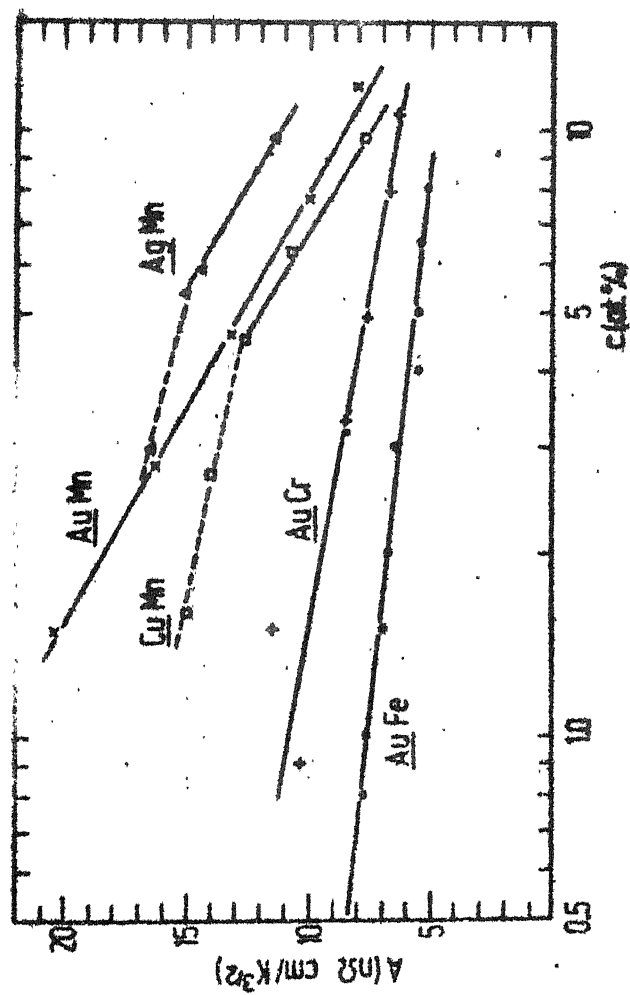


Figure 9 : Coefficient of the  $T^{3/2}$  dependence  $A$  ( $n\Omega\text{cm}/K^{3/2}$ ), plotted against  $\log_{10} c$  for  $\overline{\text{AuCr}}$ ,  $\overline{\text{AgMn}}$ ,  $\overline{\text{CuMn}}$  and  $\overline{\text{AuFe}}$  alloys (after ref. [80]).

that  $\frac{\Delta\rho}{\rho} \propto H^n$  where  $n > 1$ . At low fields  $n \simeq 2$  for all the alloys studied. The field  $H_q$  upto which the quadratic nature of  $\Delta\rho/\rho$  persists is different for different alloys. Besides at low fields,  $\frac{\Delta\rho}{\rho}$  is weakly dependent on temperature for all  $T < T_g$ .

### 1.3.5 Neutron Scattering:

The q-dependent susceptibility  $\chi(q)$  was determined by Murani and coworkers [37-39,85,86] from neutron scattering experiments using the relation

$$\frac{d\sigma}{dq} \propto k_B T \chi(q) \quad (1.5)$$

Some remarkable features emerged -

- (a) the temperature  $T_F$  where the peak in  $\chi(q)$  occurs was always much higher than the temperature corresponding to the cusp in a.c. susceptibility, as shown in Figure. 10.
- (b) the freezing temperature  $T_F$  is q-dependent-higher the q-value lower is  $T_F$  as shown in Figure 11.
- (c) there is a distribution of relaxation times.

### 1.4 Theories of Spin Glasses:

Mydosh [87] commented that it is easier to discover a new SG material than to understand the behaviour of those already known. The nature of the SG transition in even the simplest systems e.g. AuFe and CuMn is controversial. In this field of research, as in many other branches of condensed



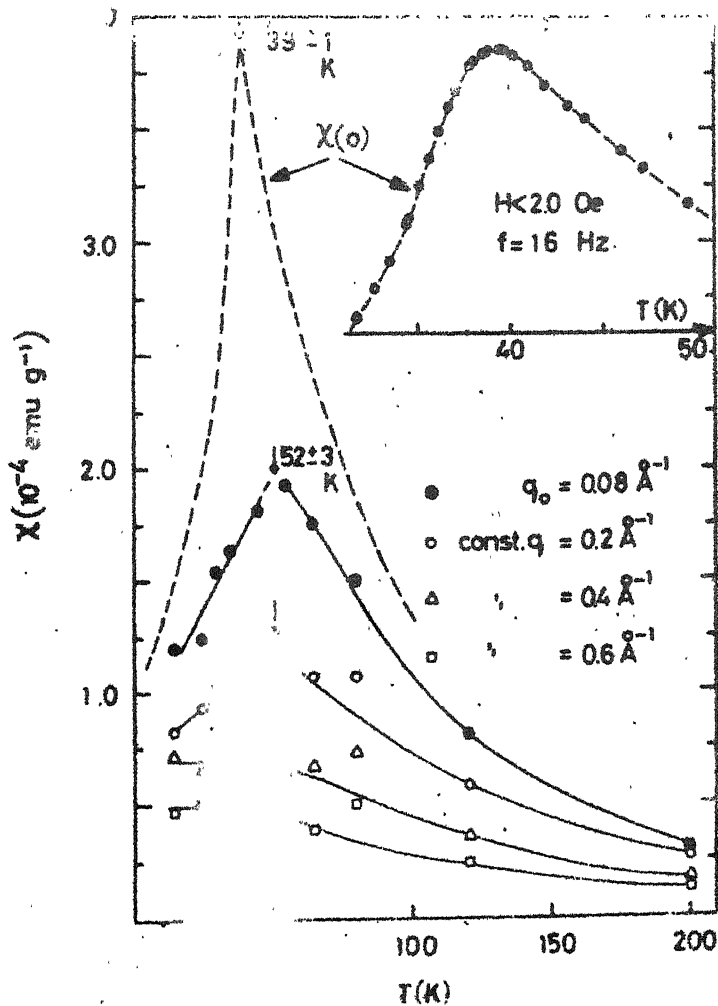


Figure 10 : The susceptibility  $\chi(q)$  versus  $T$  for Cu-8 at % Mn alloy. The dashed curve is the measured a.c. magnetic susceptibility of the same large sample as used for the neutron scattering experiment (after ref. [37]).

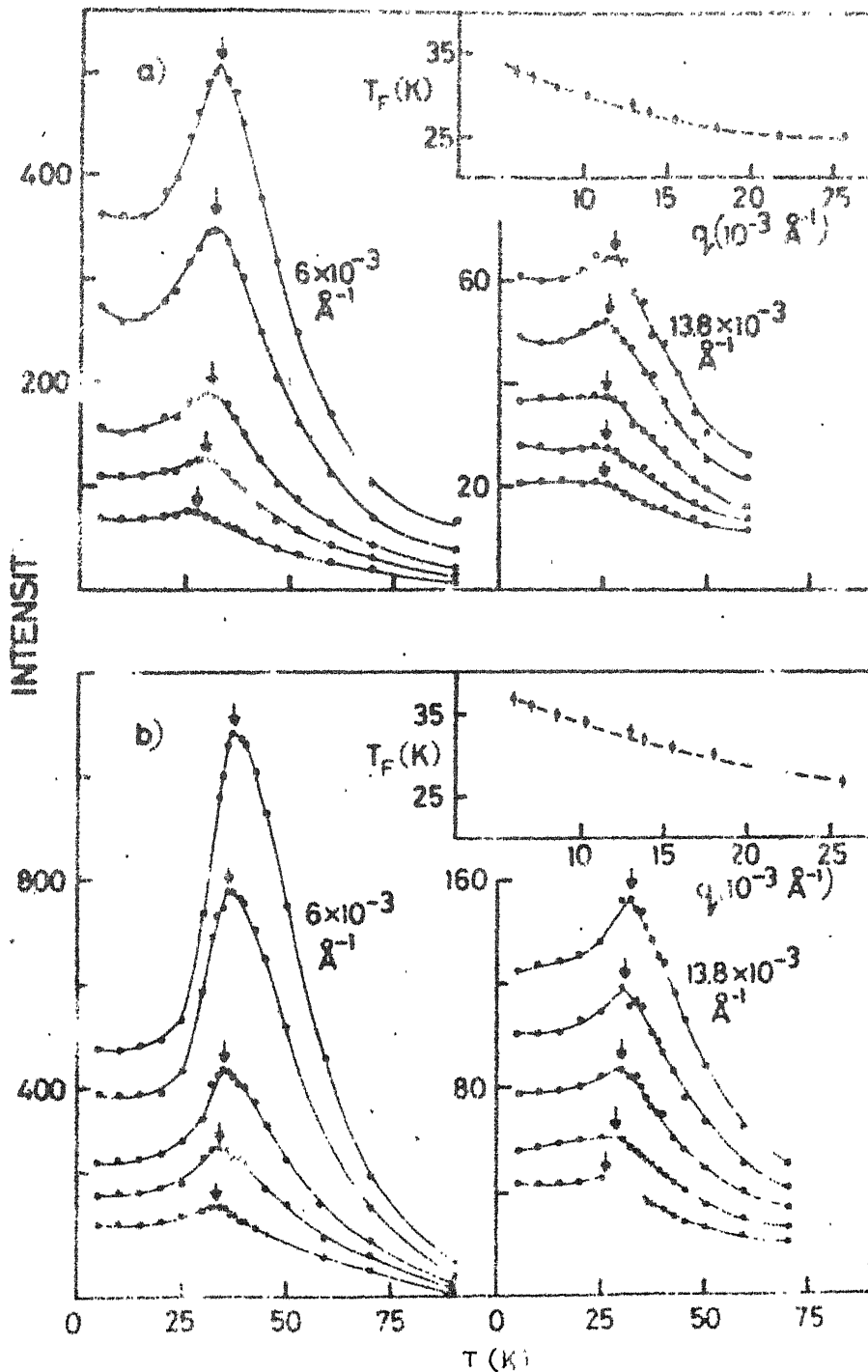


Figure 31 : Small angle neutron scattering intensity as a function of the temperature for a series of  $q$ -values for AuFe alloys containing (a) 10 at % Fe and (b) 13 at % Fe. The arrows mark the temperatures of the discontinuity in the scattering which are shown as function of  $q$  in the insets (after ref. [37]).

matter physics, theory is lagging far behind experimental developments. So far almost all conventional methods have had limited success. The solution may require the introduction of some totally new concepts. Many new ideas have already been introduced (e.g., frustration) and many more have been borrowed from other fields, e.g. the ideas of replica and long range order in time borrowed from polymer physics, concepts of gauge invariance and spontaneous symmetry breaking from quantum field theory. Yet the whole picture, to this date, remains unclear. Moreover, a theoretician must also decide whether all kinds of SG can be explained by some general universal theory or different classes of materials showing the SG transition need different theoretical treatments. At present we can only say that the situation is not ripe for reaching any final conclusion.

In any microscopic theory in physics one has to know the nature of the interaction. Before developing a theory of SG one has to construct a reliable model for the form of the impurity-impurity interaction. Here one once again comes across an age old problem in the theory of magnetism. The Heisenberg Hamiltonian

$$\mathcal{H} = - \frac{1}{2} \sum_{ij} J_{ij} \underline{S}_i \cdot \underline{S}_j \quad (1.6)$$

is a good description of the system provided there exist 'good' local moments at the impurity sites. Since we have already assumed the existence of such moments in article 1.1

while defining a SG, one can work with either the Heisenberg model or the simpler models e.g. XY model or Ising model. But the definition of SG in article 1.1 is only a clever trick to bypass the question - "can there exist situations where the SG has to be described in terms of itinerant model [88]?" Coles [89] has shown that there exist such situations e.g. AuCo, RhFe, RhCo, etc. These are called Stoner glass to distinguish them from the ones defined in article 1.1. The defining criterion for a Stoner glass is that the SG transition temperature  $T_g$  is much less than the Kondo temperature  $T_k$ , i.e.  $T_g \ll T_k$ . Hertz [90] has developed the theory of Stoner glasses. But in this thesis we shall not discuss them further.

Next, one has to know the nature of the exchange interactions  $J_{ij}$  between the  $i$ th and  $j$ th spins. The nature of this exchange interaction, i.e. whether direct or indirect, RKKY or superexchange, has to be decided by experiment. Of course, the essential feature of this exchange is the competition between ferromagnetic and antiferromagnetic interactions, as defined in article 1.1. The dominating interaction in canonical SG with which we are primarily interested is RKKY [91-94] in nature. But its explicit form has been questioned from time to time. It was De Gennes [95] who first showed that in a disordered system the expression for the effective indirect exchange is

$$J_{ij} = \frac{J_{sd}^2}{\epsilon_F} \frac{(k_F a_o/2)^6}{\pi^3} \left[ \frac{2k_{Fij} \cos(2k_{Fij} R_{ij}) - \sin(2k_{Fij} R_{ij})}{(2k_{Fij} R_{ij})^4} \right] e^{-R_{ij}/\delta} \quad (1.7)$$

where  $\delta = \frac{\hbar k_F \tau}{m^*}$  is the mean free path of the conduction electrons and arises due to the effect of disorder,

$$a_o^3 = \text{volume of unit cell, and}$$

$$\epsilon_F = \frac{\hbar^2 k_F^2}{2m^*}$$

Later similar explicit analytical expressions were derived by Kaneyoshi [96] and Poon [97] independently, which were shown to be equivalent mathematically by Kaneyoshi [98]. Very recently de Chatel [99] has re-examined the validity of De Gennes' expression and concluded that the latter is a good approximation when the disorder is weak thereby confirming Kaneyoshi's conjecture based on model calculations that as mean free path becomes comparable with the average coherence length of disorder, Fermi energy approaches the mobility edge implying more and more deviation from De Gennes' approximation (called "factorization approximation" by de Chatel).

Since the exchange interaction in SG is random due to the quenched disorder of the system one has to assume or derive the distribution of the relevant random variable (either the random exchange interaction or the random local field) and has to invoke the idea of spatial ergodicity [100], common feature of all theories of disordered systems.

So far as the configuration averaging is concerned, random magnetic systems are more complicated than the random non-magnetic systems, because of the added degrees of freedom-spin. This causes a difficulty that can be avoided by introducing some mathematical tricks which, in turn, introduce many more problems whose solutions are still far from satisfactory. In random magnetic systems one has to carry out two averages - the thermal and configuration averages. So, the next obvious question is which should be carried first? In other words, what physical quantity should be configuration-averaged-partition function, free energy, magnetization or specific heat etc? It was Brout [101] who, in his work on random ferromagnetism, answered this question clearly for the first time. In annealed systems the "impurity" degrees of freedom  $\{I\}$  are in thermal equilibrium with other degrees of freedom, e.g. spin  $\{S\}$ , the variables  $\{S\}$  and  $\{I\}$  can be treated on the same footing and therefore the partition function  $Z$  has to be averaged over all configurations. On the other hand, in a quenched system at a low temperature  $T$  the spin degrees of freedom  $\{S\}$  are in thermal equilibrium whereas the impurity configuration  $\{I\}$  is the equilibrium configuration corresponding to a very high temperature. Therefore  $\{I\}$  cannot be treated on equal footing with  $\{S\}$ . In quenched systems one first calculates the free energy corresponding to a given configuration and then averages it over all impurity configurations.

#### 1.4.1 Theories of First Kind

These are the oldest theories of SG alloys and are based on the idea of a distribution of random local magnetic fields. The first among these that explained the low temperature linear specific heat was that proposed by Marshall [102]. He assumed (a) that the spins interact via RKKY interaction and (b) that the spins are Ising spins ( $S = 1/2$ ). He estimated the probability  $P(H,T)$ , the probability of local field  $H$  at temperature  $T$ , in an intuitive manner as discussed below: the local field distribution for ferromagnetic or antiferromagnetic systems are  $\delta$ -function in Weiss molecular field approximation but broaden out in the improved Bethe-Peierls-Weiss (BPW) approximation. In case of dilute alloys one would expect it to broaden and flatten out more so that the width of the peaks become comparable to mean magnitudes of the local field itself. Following Kittel's suggestion that this problem is closely analogous to the computation of "the line shape for NMR on dilute sample of nuclear spins distributed randomly in a crystal and coupled only by their magnetic dipole interactions", Marshall argued that the local field distribution, in analogy with NMR line shape, should also be a cutoff Lorentzian. Of course, Marshall stressed that the exact shape of the distribution is not important for explaining the temperature and concentration dependence of low temperature specific heat provided  $P(H,T)$  has a width proportional to the concentration  $c$  for dilute alloys.

The magnetic energy is given by

$$E = - \frac{1}{2} Nc \int_{-\infty}^{\infty} dH P(H,T) \mu H \tanh\left(\frac{\mu H}{k_B T}\right) \quad (1.8)$$

where  $N$  = number of sites, and the factor  $1/2$  ensure no double counting. Hence, the specific heat is given by

$$\begin{aligned} C_m(T) = & \frac{1}{2} Nc \int_{-\infty}^{\infty} dH P(H,T) \left(\frac{\mu^2 H^2}{k_B^2 T^2}\right) \text{sech}^2\left(\frac{\mu H}{k_B T}\right) \\ & - \frac{1}{2} Nc \left(\frac{\partial}{\partial T}\right) h(T) + \frac{1}{2} Nc \int_{-\infty}^{\infty} dH \left\{ \mu H - \mu H \tanh\left(\frac{\mu H}{k_B T}\right) \right\} \\ & \frac{\partial P(H,T)}{\partial T} \quad (1.9) \end{aligned}$$

where

$$h(T) = \int_{-\infty}^{\infty} dH |H| P(H,T) \quad (1.10)$$

is the mean magnitude of  $H$ .

At very low temperatures ( $T < \frac{\mu h}{k_B}$ ) most of the spins are rigidly pinned by their local fields and with further decrease of temperature there can be no further appreciable increase in the number of such frozen spins. Since we also know that only such rigidly aligned spins make appreciable contribution to  $h$ , we conclude that the temperature variation of  $h$  is negligibly small. Therefore, the second term in equation (1.9) can be dropped. Moreover, in the first and third terms only a small range of  $H$  (from  $-\frac{k_B T}{\mu}$  to  $\frac{k_B T}{\mu}$ ) makes dominant contribution, because it is only in this range that



thermal agitation dominates over magnetic order. Therefore  $P(H)$  can be replaced by  $P(0)$ . In addition, for any given reasonable variation of  $P(0)$  with  $T$ , the third term is negligible compared to the first. Hence, we finally get, after a change of variable

$$C_m(T) \approx \frac{1}{2} Nc P(0,0) \frac{k_B T}{\mu} \int_{-\infty}^{\infty} dx x^2 \operatorname{sech}^2 x \quad (1.11)$$

and hence the linear behaviour in  $T$ .

Later Marshall's intuitive theory was put on a rigorous basis by Klein and Brout [103] (KB). Their main contribution was the derivation of the local field distribution from very basic considerations. They confirmed Marshall's intuitive suggestion that the local field distribution should be Lorentzian. Unfortunately, Lorentzian is not a very well behaved distribution in the sense that all its higher moments diverge, thereby implying, in case of SG, infinite energy per spin. KB realised this drawback. They showed that this difficulty can be removed by paying attention to the real physical situation where there exists a lower cutoff in the relative distance between two impurities because the lattice structure is discrete. When the latter fact was properly taken into account the local field distribution turned out to be Gaussian which still has its width proportional to the concentration  $c$ , which was the only essential feature that Marshall assumed.

The maximum observed in the susceptibility was explained by KB as follows: first they showed that there exists a characteristic correlation length  $R_c$  beyond which  $\langle S_0 S_j \rangle = 0$  such that the "impurity spins are fully or partially correlated to the spins at the origin if they are located within radius  $R_c$  and are approximately randomly oriented if they are located outside of  $R_c$ ". Thus the solid is made up of small clusters of impurities. As the temperature is raised starting from very low temperature two competing phenomena take place - the clusters get randomized resulting in a decrease in the susceptibility, and clusters breaking up thereby increasing the susceptibility. At low temperatures the latter dominates whereas at high temperature the former dominates. As a result there occurs a maximum in the susceptibility. We shall not discuss the quantitative details of the above qualitative idea, firstly, because zero field susceptibility exhibits a sharp cusp rather than a maximum, and secondly, because the physical picture used by KB is contradictory to reality as will be shown very soon.

Klein [104] discussed quantitatively the nature of the local field distribution and its variation with concentration and temperature. Since the increase of concentration results in a decrease of  $R_c$ , the effect on  $P(H)$  is "to remove some probability from the central portion of  $P(H)$  and to place it into the wings". Unfortunately, the variation of  $P(H)$  with temperature turned out to be unphysical - the most probable field shifts to higher values as the

temperature is raised. This is so because of the following reason: a spin situated in between two others can screen off the interaction. Higher temperature implies weaker screening and hence higher effective field. This is contrary to Marshall's qualitative conclusion: "the effect of correlation (at lower temperature) must be to reduce the energy and this can only be done by arranging for spins to sit in fields of larger magnitude".

The possible source of error in Klein's theory was pointed out by Liu [105]. He stressed that the screening effect, if present at all, will be complete only at 0°K. At any finite temperature screening may be negligibly small and hence KB approximation breaks down. Klein extended the formulation to include the effect of external magnetic field [106] and to explain the high temperature properties [107].

One interesting feature of the internal field distribution  $P(H,T)$  is that it obeys the scaling law [108]

$$P(H,T) = \frac{1}{c} f\left(\frac{H}{c}; T/c\right) \quad (1.12)$$

for  $c \rightarrow 0$ .

Some other scaling laws follow from this form, e.g.

$$\begin{aligned} \chi(T) &= K(T/c) \\ \frac{\Delta C_p}{c} &= \Gamma(T/c) \end{aligned} \quad (1.13)$$

Such scaling laws are consequences of the power law behaviour of the form  $1/r^n$  of the interaction. One may wonder what is

the use of these scaling laws. The beauty of these laws lies in the fact that  $K(x)$  and  $\Gamma(x)$  being concentration independent functions, properties for all concentrations can be derived from those of a single concentration for any given system, e.g. AuFe. RKKY interaction has an additional unique feature that the exponent  $n=3$ =space dimensionality of the system. This feature leads to another simplification and universal behaviour - the properties of all the canonical SG systems for all concentrations can be derived from those of a single system for a single concentration. An excellent discussion on these scaling laws can be found in Rammal and Souletie [15].

It appears that the success of the theories of first kind was the explanation of low-temperature linear specific heat. But this explanation is also not very satisfactory [109]. One does not get linear specific heat for Heisenberg spins in the same theoretical framework, because

$$P(H) \propto H^2$$

However, Rivier [110] counterargued that "the relevant quantity, however, is the finite  $P(H_z)$  rather than  $P(H)$ , because the local magnetic energy  $\underline{S} \cdot \underline{H}$  involves a scalar product, hence a preferred direction (that of  $\underline{S}$ )", i.e.,

$$\begin{aligned} E &= \int dH \quad P(H) \quad \underline{S} \cdot \underline{H} \\ &= \int dH_z \quad P(H_z) \quad H_z B(H_z, T) \end{aligned} \quad (1.14)$$

where  $B(H_z, T)$  is the Brillouin function for the spin  $S$  in a field  $H_z$ . Anderson [11] rejected this argument saying "confusion on this point is widespread....it is quite clear that  $\underline{\mu} \cdot \underline{H} = \mu H$ ". The latter situation arises because, in equilibrium,  $\underline{\mu}$  is parallel to  $\underline{H}$ . We shall see an alternative explanation of linear specific heat in article 1.7.2. More about the local field distribution will be discussed later.

The drawbacks of this kind of theories are summarized below:

- (a) they predict a rounded maximum in zero field susceptibility  $\chi$  instead of a sharp cusp,
- (b) the magnitude of  $\chi(T)$ , contrary to experiment, is independent of concentration, to first order (i.e., for low concentration),
- (c) that the peaks in  $\chi(T)$  are independent of fields (for low fields), contrary to experiment,
- (d) that the temperature of the maximum should be directly proportional to concentration whereas experiment shows weaker-than-linear dependence.

These points had been cited by Cannella [112] as failures of these earliest theories.

### 1.4.2 Theories of Second Kind

Though the theories included in this category may differ from each other in their quantitative details, they share a common feature, viz., they show SG transition as a genuine magnetic phase transition. There are five steps in the study of a phase transition:

- (i) identification of the relevant physical characteristic(s) that distinguishes the two phases of the system,
- (ii) quantification of the distinguishing characteristic(s) found in step (i) by defining order parameter(s),
- (iii) developing a mean field theory (MFT) in terms of the free energy as a function of the order parameter,
- (iv) verification that in the long-range limit (i.e., infinite range model) MFA gives exact answer,
- (v) improvement of MFT results by incorporating critical fluctuations in the theory e.g. by the method of renormalization group.

Unfortunately, in the case of SG none of these steps have yet been completed very satisfactorily. However, we shall follow this scheme in our discussion of the SG 'phase transition'.

### Steps I and II:

Identification of the relevant order parameter is very crucial, because a system can exhibit several phase transitions characterized by different order parameters. We know that spontaneous magnetization is a measure of long-range ferromagnetic order whereas staggered magnetization is a measure of long range antiferromagnetic order. Hence, spontaneous magnetization and staggered magnetization distinguish ferromagnetic and antiferromagnetic states from paramagnetic one. In other words, spontaneous magnetization and staggered magnetization play the role of order parameters for the corresponding phase transitions. After Cannella and Mydosh's classic observation of a sharp cusp in the low-field a.c. susceptibility physicists tried to explain it as due to the onset of antiferromagnetic order. But it could not be fitted within the existing ideas of long-range antiferromagnetic order because

- (a)  $\chi_{11}$  and  $\chi_{\perp}$  below the cusp temperature  $T_g$  were different for antiferromagnetism but identical for SG,
- (b) the cusp was very sensitively field dependent, unlike antiferromagnetism,
- (c) neutron scattering [113] showed conclusively absence of any long-range order.

Therefore it was obvious that SG is distinct from ferromagnetic or antiferromagnetic phases and requires new order parameter. Edwards and Anderson [114] introduced, for

SG, an order parameter  $Q$ , now popularly known as Edwards-Anderson (EA) order parameter, defined as

$$Q = [ \langle S_i \rangle^2 ]_{av} \quad (1.15)$$

where  $\langle \rangle$  denotes thermal-average and  $[ ]_{av}$  denotes configuration average. In order to describe the dynamics of the system they introduced [15] a dynamic order parameter

$$Q(t) = [ \langle \underline{S}_i(t) \cdot \underline{S}_i(0) \rangle ]_{av} \quad (1.16a)$$

Notice that  $q(t)$  is always positive - it relaxes to its static value for  $t \rightarrow \infty$  only via values all greater than zero. The physical significance of EA order parameter is very important. In Anderson's own words [6] "we decided that since we believed, there was no LRO in space, we had to introduce a LRO in time". So, the dynamic order parameter  $Q(t)$  describes on-site autocorrelation in time, i.e., the probability that the spin at a particular site will be 'oriented' in a particular direction, at time  $t$ , if it was 'oriented' in the same direction at time  $t=0$ . This is, of course, a classical interpretation. In quantum mechanical language,  $Q(t)$  describes the probability that the spin at a particular site will be an eigenstate if it was in the same eigenstate at  $t=0$ .

Unfortunately, the choice of the order parameter, for SG phase, is not universally agreed upon. Recently,



Khurana and Hertz [116], from a study of order parameter susceptibility concluded the insufficiency of the single EA order parameter  $Q$  to describe the SG phase. They also indicated the possibility of introducing generalized order parameters involving higher order cumulants. Sompolinsky [117] introduced two order parameters  $Q$  and  $\Delta$ , to be discussed later. Plefka [118] utilized the generalized order parameters

$$Q_r = \frac{1}{N} \sum_i m_i^r \quad (1.17)$$

Parisi [119,120], in yet another context, remarked that "it is quite likely that an infinite number of order parameters is needed in the correct treatment". Parisi [121] introduced a generalized order parameter  $Q(x)$  where the variable  $x$  is defined over the interval  $[0,1]$ .

Suzuki and Miyashita [122] have discussed in detail different kinds of definitions of order parameters for SG. They, however, defined an order parameter and many of the earlier order parameters turned out to be special cases of this order parameter.

Binder and Stauffer [123] proposed to analyze the nature of the ordering in SG and relaxation from one degenerate ground state to another in terms of some new order parameter. Let the set of phase factors  $\phi_1^{(1)}, \phi_2^{(1)}, \dots, \phi_N^{(1)}$ , defined by the direction of spins  $\{S_1, \dots, S_N\}$  in the  $-l$ -th ground state, define an  $N$ -dimensional space with unit vectors

$$\underline{\varphi}^{(1)} = \frac{1}{\sqrt{N}} \{ \varphi_1^{(1)}, \dots, \varphi_N^{(1)} \} \quad (1.18)$$

They thought, "it is interesting to study the "similarity" of two ground states" produced by repeated aging and quenching in a Monte-Carlo computer experiment. They defined the order parameter

$$\underline{\psi}^{(1)} = \underline{\varphi}^{(1)} \cdot \underline{X} \quad (1.19)$$

which is just the projection of the state

$$X = \frac{1}{\sqrt{N}} \{ S_1, \dots, S_N \}$$

onto the 1-th ground state defined by the unit vector

$$\underline{\varphi}^{(1)} = \frac{1}{\sqrt{N}} \{ \varphi_1^{(1)}, \dots, \varphi_N^{(1)} \}$$

The question whether the 1-th ground state has a non-zero order parameter with respect to m-th ground state is described by whether or not

$$\cos \theta_{1m} = \underline{\varphi}^{(1)} \cdot \underline{\varphi}^{(m)}$$

vanishes. By starting from some direction  $\underline{\varphi}^{(1)}$  one can "rotate" in a higher dimensional space which physically implies relaxation of the system from one ground state to another. Notice that  $\underline{\psi}^{(1)}$  looks at a particular configuration of all the spins whereas the order parameters defined earlier look at a single spin averaged over different configurations.

Kirkpatrick and Young [124,125] discussed the drawbacks of the usual definition of EA order parameter for their computation on a finite lattice. An external field, however small, was required to break the symmetry in order to generate a non-zero  $\langle S_i \rangle$  in that particular direction and, then, after taking the thermodynamic limit, one can put the external field = 0. They estimated that  $\langle S_i \rangle$  is nonzero for  $h \gg h^* = \frac{T}{\sqrt{N}}$  and hence  $h^* \neq 0$  in finite lattice. One should, however, remember that the field applied  $h_i = \epsilon_i h$  with  $\epsilon_i = \pm 1$  with equal probability so that the process of symmetry breaking differs from that in a ferromagnet where a uniform field is used. The utility of the symmetry breaking lies in the fact that only states in the vicinity of one of the ground states are included in the ensemble average. In case of large but finite systems Morgenstern and Binder's [126] order parameter

$$\lim_{R_{ij} \rightarrow \infty} [\langle S_i S_j \rangle^2]_{av} \quad (1.20)$$

with short range interactions is very convenient one since  $S_i S_j$  does not change sign when all spins are reversed. Young and Kirkpatrick generalized this order parameter to infinite-range model

$$Q^{(2)} = [\langle S_i S_j \rangle^2]_{av} \quad (i \neq j) \quad (1.21)$$

The superscript (2) suggests that " $Q^{(2)}$  is something like  $Q^2$  but not exactly equal to this".

Adkins and Rivier [127] assumed that the local magnetization, a short range order parameter, describes the SG phase. This differs from EA order parameter by the fact that the latter does not assume any SRO in space but only a LRO in time. Later Medvedev [128] generalized EA order parameter to include short range magnetic order by writing

$$[ \langle S_i \rangle \langle S_j \rangle ]_{av} = Q \phi_{ij} \quad (1.22)$$

where  $\phi_{ij} \neq 0$  even for  $i \neq j$ , and  $\phi_{ij} \rightarrow 0$  as  $|\underline{R}_i - \underline{R}_j| \rightarrow \infty$ .

The last condition is necessary to avoid LRO. Medvedev's order parameter reduces to EA order parameter if

$$\phi_{ij} = \delta_{ij}$$

Sompolinsky [129] suggested the introduction of staggered magnetization associated with the eigenvalue spectrum of the random exchange matrix as the SG order parameter. The staggered magnetization  $M_\lambda$  is defined by

$$M_\lambda = \sum_i \langle i | \lambda \rangle S_i \quad (1.23)$$

$\langle i | \lambda \rangle$  being the orthonormal eigenvector of the random matrix  $J_{ij}$  corresponding to the eigenvalue  $J_\lambda$ .

### Step III:

Edwards and Anderson [114] hoped that their theory will play the same role as Weiss molecular field theory did

for LRO phases. In order to average the configuration dependent free energy over all possible configurations EA utilized a technique called "replica trick". The trick converts the quenched problem into an annealed problem by using the identity

$$\ln Z = \lim_{n \rightarrow 0} \frac{1}{n} (Z^n - 1) \quad (1.24)$$

So, effectively what one averages is  $Z^n$ . For integral  $n$ ,

$$Z^n = \prod_{\mu=1}^n Z_{\mu}$$

can be interpreted as the partition function of a composite system consisting of  $n$  non-interacting replica of the system and hence the name. More details of the replica method will be discussed in the next section. EA had to modify the definitions of their order parameters in the replica approach. EA assumed

$$M_{\alpha} = [ \langle S_i^{\alpha} \rangle ]_{av} \quad (1.25)$$

and

$$Q_{\alpha\beta} = [ \langle S_i^{\alpha} \rangle \langle S_i^{\beta} \rangle ]_{av} \quad (\alpha \neq \beta) \quad (1.26)$$

as the only relevant order parameters, and neglected all other combinations including the higher order ones. At this point one should notice that Khurana and Hertz's [116] conclusion about the insufficiency of the above order parameters, as

stated in the previous section, can be anticipated here. One has to have infinite number of order parameters if the successive higher order terms including three or more spin operators are not negligible. Moreover, there does not exist any mathematically rigorous proof that the order parameters defined by equations (1.25) and (1.26) reduce to M and Q in the limit  $n \rightarrow 0$ .

EA assumed adhoc that the spins are classical spins and the exchange interactions obey Gaussian distribution. They were able to show the existence of a critical temperature  $T_g$  above which  $q$  vanishes.  $T_g$  is called the SG transition temperature. Finally they obtained the following expression for zero field susceptibility

$$\chi(T) = \chi_0 (1 - q) \quad (1.27a)$$

where

$$\chi_0 = \frac{NS (S+1)(g\mu_B)^2}{3k_B T} \quad (1.28)$$

is the susceptibility of free spins. This expression (1.27a) exhibit a cuspat  $T_g$ , as observed experimentally [ 73] . But the overall shape of the  $\chi(T)$  curve obtained by EA did not agree with experimental results. The slope of  $\chi(T)$  turned out to be zero just below  $T_g$  contrary to experiment. Fischer [ 130] attributed this discrepancy to the quantum nature of spins and developed the theory for quantum spins following EA very closely. Assuming validity of the standard

result of linear response theory (LRT), viz.,

$$X(T) = \sum_{ij} \{ [ \langle S_i S_j \rangle ]_{av} - [ \langle S_i \rangle \langle S_j \rangle ]_{av} \} x_0 / NS(S+1) \quad (1.29)$$

and definition of  $Q$ , he obtained

$$X(T) = x_0 \left[ 1 - \frac{Q}{S(S+1)} \right] \quad (1.30)$$

in the absence of SRO. He also showed that  $\frac{X(0)}{X(T_g)} = 0.69$  for  $S = \frac{1}{2}$  which agreed well with experimental result (0.6) whereas the classical theory of EA predicted a corresponding value of 0.92. Moreover in the classical limit (i.e., for classical spin). Fischer's results agreed with those of EA. Sherrington and Southern [131] developed quantum-mechanical version of EA model, independently and obtained results identical with those of Fischer [130]. Fischer [132] also extended his calculations in the presence of external magnetic field.

But, unfortunately, all was not well with EA model. It also predicted [114] a cusp in the specific heat at the same temperature  $T_g$ , which has never been observed experimentally. This is the first evidence that the SG transition, if at all a phase transition, is an unconventional one. Recently Barnes [133] showed instability of the EA ground state with respect to magnons.

Edwards [134] examined the condition for existence of SG phase by quantizing a SG consisting of randomly

arranged dipoles of moments of inertia  $I$  and experiencing random forces. SG phase was shown to exist provided the dimensionless parameter  $\frac{J_0 I}{\hbar^2}$  is greater than a critical value where the classical ground state energy is  $NJ_0$ ,  $N$  being the number of dipoles. However, physically the problem turned out to be similar to Anderson localization [135]. The latter aspect will be discussed in detail later.

Physicists felt uneasy at the use of "replica trick" because analytical continuation upto  $n = 0$  involves fractional values of  $n$  also and the physical significance of fractional replica was not clear. Sherrington [136] and Southern [137] derived expressions similar to those of Fischer using simple mean field theory in much more transparent manner without using replica trick.

The EA model has been extended to include uniform uniaxial anisotropy [138] and also random uniaxial anisotropy [139]. The latter exhibits a broadened maximum in the specific heat instead of a sharp cusp in the former but the broadened maximum occurs at almost the same temperature as the cusp.

#### Step IV:

Sherrington and Kirkpatrick (SK) [140,141] applied the EA theory to an Ising model where each of the spins are coupled to all others by infinite ranged infinitesimally weak exchange interactions. In other words, in this model



the number of nearest neighbours  $Z$  is infinity. SK used replica trick and Gaussian distribution for exchange interaction:

$$P(J_{ij}) = \frac{1}{\sqrt{2\pi}J} \exp \left[ -\frac{(J_{ij} - J_0)^2}{2J^2} \right]$$

where

$$J_0 = \tilde{J}_0/N$$

and

$$J = \tilde{J}/N^{1/2}$$

with  $\tilde{J}_0$  and  $\tilde{J}$  both intensive. The factors  $N$  and  $N^{1/2}$  were introduced to make the thermodynamic limits physically sensible. While calculating the free energy, SK interchanged the two limits  $N \rightarrow \infty$  and  $n \rightarrow 0$ . The equilibrium values of the order parameters  $M(T,H)$  and  $Q(T,H)$  are given by two coupled integral equations, called SK equations.

However, SK model suffers from two serious drawbacks:

- (i) it gives negative entropy at sufficiently low temperatures. The authors themselves speculated that "this unphysical behaviour has its origin in the interchange of limits  $N \rightarrow \infty$  and  $n \rightarrow 0$ , but the consequences are confined to low temperatures".
- (ii) the solution obtained does not correspond to a minimum of the free energy though it is an extremum.

Van Hemmen and Palmer [142], on the other hand, showed that the unphysical results appear because of nonuniqueness of

the limit  $n \rightarrow 0$ .

Since SK's original derivation is complicated we shall sketch the simpler derivation of Mookerjee [143]. This method has the advantage that no replica trick is required because the configuration averaging is done at the end of the calculation. The configuration dependent free energy is given by

$$F(\{\underline{r}_k\}) = \left(\frac{1}{2}\right) \sum_i \sum_j J(|\underline{r}_i - \underline{r}_j|) \sigma_i \sigma_j - \frac{1}{\beta} \sum_i \ln \cosh \left\{ \beta \sum_j J(|\underline{r}_i - \underline{r}_j|) \sigma_j \right\} \quad (1.31)$$

where  $\sigma_k = \langle S_k \rangle$  is the thermal average of the spin at site  $k$  and hence proportional to the local magnetization. Stationarity of free energy, i.e.

$$\frac{\partial F(\{\underline{r}_k\})}{\partial \sigma_k} = 0 \quad (1.32)$$

implies

$$\sigma_k = \tanh \left[ \beta \sum_i J(|\underline{r}_i - \underline{r}_j|) \sigma_i \right] \quad (1.33)$$

In the absence of SRO, i.e., provided the occupation probability

$$P(\underline{r}_1, \dots, \underline{r}_N) = P(\underline{r}_1), \dots, P(\underline{r}_N)$$

One gets local field distribution

$$P(h) = \frac{1}{2\pi} \int dk e^{ikh} e^{cF(k)} \quad (1.34)$$

in thermodynamic limit, where

$$F(k) = \sum_{\underline{r}} \int dh P(h) \{1 - e^{-ikK(\underline{r}) \tanh(\beta h)}\} \quad (1.35)$$

Equations (1.34) and (1.35) together constitute self consistent equations for  $P(h)$ . Expanding and retaining only terms upto the order of  $K^2$  one gets

$$F(k) = \int dh P(h) [ik K_0 \tanh(\beta h) + \frac{1}{2} k^2 K_1^2 \tanh^2(\beta h)] \quad (1.36)$$

where

$$K_0 = \sum_{\underline{R}} K(\underline{R}) \quad (1.37a)$$

$$K_1^2 = \sum_{\underline{R}} K^2(\underline{R}) \quad (1.37b)$$

Defining,

$$M = \int dh P(h) \tanh(\beta h) \quad (1.38)$$

$$Q = \int dh P(h) \tanh^2(\beta h) \quad (1.39)$$

One finally gets the local field distribution  $P(h)$  to be Gaussian

$$P(h) = \frac{1}{\sqrt{2\pi cQ K_1^2}} \exp\left[-\frac{(h - c K_0 M)^2}{2 cQ K_1^2}\right] \quad (1.40)$$

Hence, substituting (1.40) into (1.38) and (1.39) we get

$$M = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{-x^2/2} \tanh\left(\frac{T_c M}{T} + \frac{T_g \sqrt{Q}}{T} x + \frac{\alpha}{T}\right) dx \quad (1.41)$$

and

$$Q = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{-x^2/2} \tanh^2\left(\frac{T_c M}{T} + \frac{T_g \sqrt{Q}}{T} x + \frac{\alpha}{T}\right) dx \quad (1.42)$$

where,

$$\alpha = p_{\text{eff}} \mu_B H/k_B, \quad (1.43)$$

$p_{\text{eff}}$  being the effective Bohr magneton number.

The two coupled integral equations (1.41) and (1.42) are solved selfconsistently to get the equilibrium values of  $M$  and  $Q$ . This analysis led to the following conclusions:

- (a) three phases-paramagnetic ( $M=0, Q=0$ ), spin glass ( $M=0, Q \neq 0$ ) and ferromagnetic ( $M \neq 0, Q \neq 0$ ) appear in the phase diagram (Figure 12),
- (b) all the transition lines in the absence of external field are second order,
- (c) specific heat is zero at  $T = 0$  as expected from thermodynamics,
- (d) specific heat displays a cusp for transition to SG but exhibits a discontinuity for transition to ferromagnetic phase.

The temperature and field dependence of  $M$  and  $Q$  in the SG phase have been studied by solving equations (1.41) and (1.42) numerically for AuFe (6.6 at.%) and CuMn (0.7 at.%) by Mookerjee and Chowdhury [144]. Klein [145], Kaneyoshi [146] and Plefka [147] gave alternative derivations of

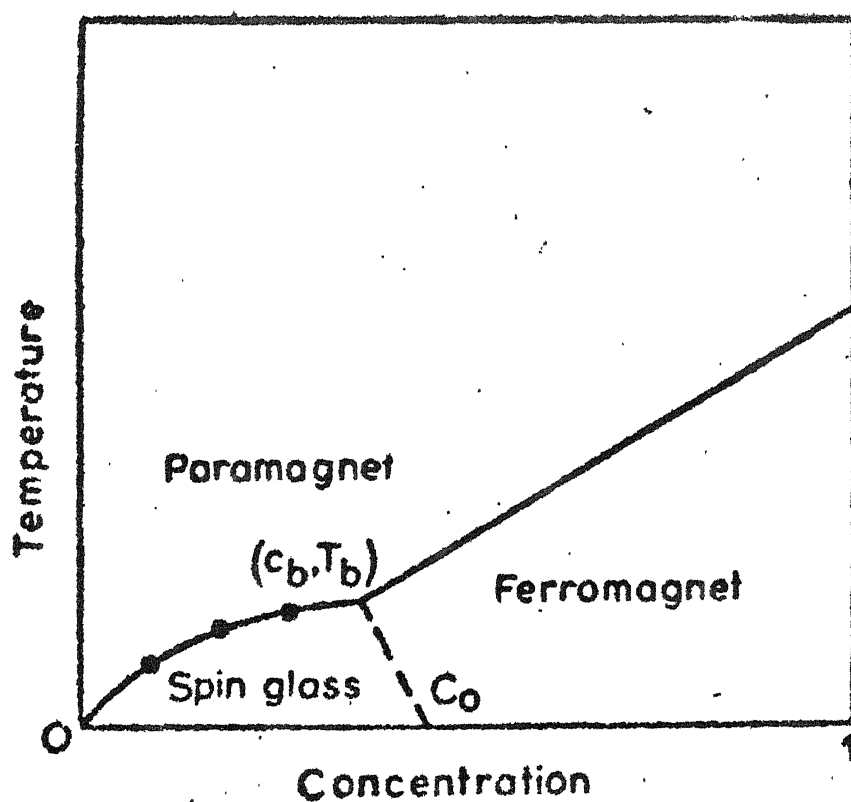


Figure 12 : Phase diagram in the temperature-concentration plane (after ref. [143]).

equations (1.41) and (1.42).

We want to stress that although the final equations (1.41) and (1.42) are the same in both Mookerjee's and SK approaches there are some fundamental differences between the two approaches. First of all, Mookerjee's model is based on RKKY interactions whereas SK used infinitely weak gaussian distributed infinite range interaction, though both are infinite ranged. Secondly, Mookerjee's approach is not based on any replica trick and so the interchange of the two limits  $N \rightarrow \infty$  and  $n \rightarrow 0$  is not required. The configuration averaging is done at the end of the calculation. The free energy is a true minimum with respect to single spinflips as computed before configuration averaging. But the stability analysis against arbitrary numbers of simultaneous spinflips has not been investigated. The entropy before configuration-averaging is positive definite and hence expected to remain so even after averaging.

All the above MFT predict  $T_g \propto c^{1/2}$ , and scaling laws support  $T_g \propto c$  whereas experimentally observed exponent varies between wide ranges. Explanations for deviation from the MF predictions were given by Larsen [148]. He took account of the facts that the conduction electrons responsible for s-d interaction may have finite mean free path due to

- (i) self damping, i.e., scattering by the localized spins  
( $\lambda \propto \frac{1}{c}$ )
- (ii) scattering by other defects ( $\lambda$  independent of concentration).

Besides, the fluctuation in the nearest neighbour distance also adds to the deviation from MFT prediction.

It is very interesting to note that Japanese groups worked out, independently, and almost simultaneously with EA, the MFT of glass-like phase (GLP) [149-152] and random ordered phase (ROP) [153-156] for short as well as long range interactions. Both GLP and ROP later turned out to be equivalent to MFT of SG. Takano [157] and Sakata et.al. [152] compared GLP with ROP. GLP is characterized by  $[\langle S_i \rangle]_{av} = 0$  and  $[\langle S_i \rangle^2]_{av} \neq 0$  whereas ROP is characterized by  $[\langle S_i \rangle]_{av} = 0$  and  $[\langle S_i \rangle^2]_{av} \neq 0$  and hence are qualitatively equivalent. The differences in the quantitative results is due to the different mathematical approximations adopted.

De Almedia and Thouless [158] first showed that the SK solution does not correspond to the minimum of the free energy and they concluded that the permutation symmetry between replicas must be broken in the "SG phase". The Almedia-Thouless instability turned out to be a special case of Pytte-Rudnick instability [159].

So, one feels uneasy at this point because the authors of references [140], [142] and [158] blame different aspects of the SK model for the disease. Moreover, Kosterlitz et.al. [160] showed that for the spherical model identical results are obtained with and without use of replica trick.

Khurana and Hertz [116], in an alternative (mathematically equivalent) diagrammatic approach, without using replica trick, showed that this instability arises due to the insufficiency of the order parameter  $Q$  to describe the SG below  $T_g$ . In this approach, the instability is signalled by a negative value of the order parameter susceptibility  $D$  below  $T_g$ , where  $D$  is defined by

$$Q = D h^2,$$

$h^2$  being the mean square value of random field  $h_j$ . For stability  $D$  must be positive definite. Unlike the prescription of breaking the replica symmetry, Sompolinsky [117], Hertz et.al. [161] and Hertz [162] suggested dynamics as a cure for this instability. We shall first discuss replica symmetry breaking schemes and then the dynamical approach.

Blandin et.al. [163], Parisi [119-121] and Bray and Moore [164-166] suggested different schemes of replica-symmetry breaking. Here we shall discuss Parisi's scheme which is the most popular among all.

Parisi [119-121, 167-171] suggested the following parametrization

$$\begin{aligned} Q_{\alpha\alpha} &= 0 \\ Q_{\alpha\beta} &= Q_i \quad \text{if } I(\alpha/m_i) \neq I(\beta/m_i) \text{ and} \\ &\quad I(\alpha/m_{i+1}) = I(\beta/m_{i+1}) \end{aligned}$$

where  $I(x)$  is an integer valued function, i.e.  $I(x)$  is the smallest integer equal to or greater than  $x$ .  $Q_i$  ( $i=0, K$ )



are real numbers and  $m_i$  ( $i=1,K$ ) are integral numbers such that  $\frac{m_{i+1}}{m_i}$  is an integer ( $i \geq 1$ ). Then putting in the basic requirements (i) that the configuration-averaged free energy be finite, (ii) that it be a minimum with respect to  $Q$  and (iii) that its minimum at high temperature be located at  $Q = 0$ , he obtained

$$\chi(T) = \frac{1}{T} \int_0^1 [1 - Q(x)] dx \quad (1.27b)$$

after taking the limit  $K \rightarrow \infty$  so that the order parameter  $Q(x)$  is now a function of  $x$  defined over the interval  $[0,1]$ . From numerical calculations for  $K = 1, 2$  etc., and then extrapolating to  $K \rightarrow \infty$  Parisi [168] obtained

$$\chi(T) = \frac{1}{T} \left[ (1 - \bar{x}) (1 - Q_{\max}) + \int_0^{\bar{x}} \{1 - Q(x)\} dx \right] \quad (1.27c)$$

where,

$Q(x)$  is linear in  $x$  for  $x \leq \bar{x}$ , and

$Q(x) = Q_{\max} = Q_{EA}$  for  $x > \bar{x}$ .

This abstract scheme has been explained pictorially by Rammal and Souletie [15].

There is, of course, lot of arbitrariness in this scheme so far as the choice of the values for the various parameters are concerned. However, Parisi [168] showed that entropy becomes less and less negative as  $K$  is increased from zero (no symmetry breaking) to higher and higher values (severe symmetry breaking). Therefore, one expects that the best possible choice for the parameters should correspond to

the most severe symmetry breaking among the replicas. Thouless et.al. [172] and De Dominicis and Kondor [173] showed that Parisi's solution is marginally stable.

Though the meaning of  $x$  was not clear from the original formulation, Gabay and Garel [174] interpreted it in the light of two fluid model of superfluidity as follows: 'a fraction  $\bar{x}(T)$  of the spins has condensed and a fraction  $1-\bar{x}(T)$  remains normal. This condensed state spans all  $x$  ( $0 < x < \bar{x}$ ), so that  $x$  appears as the probability that a given spin has an EA order parameter lying in the interval  $(0, Q(x))$ '. The condensed part consists of overblocked plaquettes, forming 'walls', whereas the normal part are frustrated forming 'domains'. In this light "SK solution can be viewed as a solution without walls". The hint for this interpretation came from the fact that  $\bar{x} = \frac{1}{2}$  at  $T = 0$  predicted by Parisi Toulouse hypothesis, to be discussed later. Parisi's order parameter  $Q(x)$ , as discussed above, is a function of one variable  $x$  ( $0 \leq x \leq 1$ ). Recently Jonsson [175] presented a scheme whereby the order parameter becomes a function on a unit square, i.e.,  $q(x,y)$ , ( $0 \leq x \leq 1$ ,  $0 \leq y \leq 1$ ). However, physical interpretation of  $x$  and  $y$  is not yet clear. Later on Sompolinsky [117] related  $x$  to the spectrum of relaxation times. We shall discuss Sompolinsky's approach later in this article.

Very recently Parisi [176] and Houghton et.al. [177] have suggested interpretation of  $x$  and  $Q(x)$  in terms of the probability distribution of the overlap of the magnetization in different pure states of the system.

Now let us discuss the alternative (dynamic) approach suggested by Hertz et.al. [161] in order to remove the instability. Using a diagrammatic expansion, they showed that in the limit  $w \rightarrow 0$  the self consistent equation for the dynamical response function  $\bar{\chi}(w)$  has two solutions - the solution consistent with fluctuation-dissipation theorem is the same as the usual equal time spin-spin correlation function of the static formalism and leads to instability; the second solution, on the other hand, though violates fluctuation-dissipation theorem (FDT) stabilizes the ground state against order parameter fluctuations and yields a finite positive value of the order parameter susceptibility.

Next we shall discuss Sompolinsky's approach and then correlate the works of various authors as proposed by Hertz [162].

Sompolinsky [117] investigated the static (statics as a limit of dynamics) properties of SG using a dynamic approach. He assumed that there exist a distribution of relaxation times, instead of a single one, all of which become infinite in the thermodynamic limit. In order to correlate with Parisi's results, he parametrized the relaxation times  $t_x$  in a decreasing order by an arbitrary parameter  $0 \leq x \leq 1$  such that

$$Q(x) = \overline{[ \langle S_i(0) S_i(t_x) \rangle ]}_{av} \quad (1.16b)$$

measures the correlation retained at time scale  $t_x$ . The maximum value

$$Q(1) = Q_{EA}$$

The most interesting result was that equilibrium value  $\chi(0)$  of susceptibility was given by

$$\chi(0) = \beta [ 1 - Q_{EA} + \Delta(0) ] \quad (1.27)$$

whereas the quasi equilibrium value  $\chi(1)$  after a time large compared to microscopic times but small with respect to macroscopic time scales was given by Fischer's relation  $\chi(1) = \beta (1 - Q_{EA})$ .  $\Delta(x)$  is an additional order parameter:

$$\Delta(x) \longrightarrow 0 \text{ as } x \longrightarrow 1$$

Also  $Q(x) \rightarrow 0$  as  $x \rightarrow 0$ .

But a peculiar feature was that "since the scale of  $x$  is arbitrary the theory can only establish a relation between  $Q(x)$  and  $\Delta(x)$  but cannot determine both of the functions, except at the uniquely defined end points 0 and 1" [117].

Sompolinsky free energy functional was rederived by de Dominicis et.al. [178] by the replica approach in a static treatment. This was another indication of a possible close relation between Parisi's replica symmetry breaking scheme and Sompolinsky's dynamic approach. Further support for such close relation came from the observation [179] that the free energy could be written as the solution of a differential equation—one of the special cases, viz.

$$\Delta'(x) = -xQ'$$

yields Parisi's differential equation [168]. This relation was also obtained by Sompolinsky, because as already stated,

his theory could not determine  $Q$  and  $\Delta$  independently but established only the relation between them.

Hertz [162] showed later that the SK spin glass is only in dynamic equilibrium. He developed the dynamical theory for a finite system. Since no finite system can exhibit a phase transition, he assigned a finite width  $\varepsilon$  to the  $\delta$ -function part in the spin correlation function  $C(w)$ . The spin correlations die out for  $t \gg \varepsilon^{-1}$  ( $\varepsilon \rightarrow 0$  as the size of the system  $\rightarrow \infty$ ) and susceptibility becomes identical with that of free paramagnetic spins. Thus Almeida-Thouless instability is suppressed only for time scales  $t \ll \varepsilon^{-1}$ . Of course, for infinite systems, which are relevant for comparison with experiments, this theory remains dynamically stable for all times because  $\varepsilon \rightarrow 0$ . Nevertheless, no stable equilibrium description is provided. Hertz interpreted this in the light of the large number of metastable states in SG and will be discussed later.

In addition, assuming that equilibration proceeds in a stepwise manner — successively larger and larger spin clusters flipping at successively longer and longer time scales — Hertz was able to interpret Parisi's  $Q(x)$  as "a measure of the local persistent spin autocorrelation for a metastable state where equilibrium has been reached with respect to all flips of spin clusters upto a size  $(1-x)N$ , but not for flips of larger clusters" [180].

Although the mathematical structure of this theory looks identical with that of Sompolinsky, there is a crucial difference — no explicit adhoc violation of fluctuation-dissipation theorem is necessary though such a violation was concluded by Sompolinsky [117], and by Hertz et.al. [161].

Recently Bray [181] examined the effect of including a site-dependent magnetic field proportional to the eigenvector with largest eigenvalue of the random matrix  $J_{ij}$  and concluded that it only suppresses strongly, but cannot remove Almedia-Thouless instability.

#### Parisi-Toulouse (PaT) Hypothesis:

PaT hypothesis [182-184] (also called projection hypothesis) states that in the SG phase the entropy does not vary with the magnetic field, i.e.,

$$S(T,H) = S(T)$$

implying that equilibrium magnetization

$$M(T,H) = M(H)$$

which together with the hypothesis

$$Q(T,H) = Q(T)$$

leads to  $F(T,H) = F_1(T) + F_2(H)$ .

The predictions of this hypothesis agree well with results of monte-carlo simulations [184]. If extrapolation to infinite system is allowed, Ma and Payne's [185] numerical computation

of entropy on a finite system with long ranged interactions justifies PaT hypothesis. However, PaT hypothesis has been shown to be approximate [186].

### Nonlinear Susceptibilities:

If paramagnet-to-SG is a genuine equilibrium phase transition then  $\chi_2$  should diverge at  $T_g$  with a universal critical exponent. However, observed variation of this exponent with temperature led Fähnle and Egami [187] to the conclusion that the divergence of  $\chi_2$  "has nothing to do with a real critical phenomenon". We shall not discuss the nonlinear susceptibilities any more in this thesis.

### Improved Mean Field Theories:

The Bethe approximation for short range site random as well as bond random Ising model has been carried out by Katsura et.al. [188,189], Ghatak and Moorjani [190], Klein et.al. [191], Morita [192] and Horiguchi [193]. An attempt to cure the diseases of the SK model was made by Thouless et.al. (TAP) [194] by applying a diagrammatic method and without using the replica trick. They observed that "it is simpler, and perhaps more physical, to observe that the set of diagrams contributing in order  $N$  are just those which would remain on a "Bethe lattice" or Cayley tree, all diagrams with no loops. Any diagram containing a closed ring is necessarily of order  $N/2$  or less, since the internal

connection reduces the number of site summations by one". Moreover the Ising model on Bethe lattice is in principle exactly soluble by the so called 'Bethe' or 'cluster' or Bethe-Pierls-Weiss' (BPW) method. The final equations for local magnetization, known as TAP equation, is

$$M_i = \tanh (\beta h_j) \quad (1.44)$$

where

$$h_i = \sum_j J_{ij} M_j - \beta \sum_j J_{ij}^2 (1 - M_j^2) M_i \quad (1.45a)$$

The physical interpretation of equation (1.45a) is very clear - it subtracts out the Onsager reaction field from the local magnetic field acting on a spin. This can be seen as follows: the field created by  $i$ th site at the  $j$ th is  $J_{ij} M_i$  and hence induces a magnetization  $J_{ij} M_i \chi_{jj}$  where  $\chi_{jj}$  is the local magnetic susceptibility at the  $j$ th site. This induced moment produces a reaction field  $J_{ij}^2 M_i \chi_{jj}$  at the site  $i$ . Summing over all  $j$ 's, the total reaction field at site  $i$  is  $\sum_j J_{ij}^2 \chi_{jj} M_i$  and hence the reaction field  $= \beta \sum_j J_{ij}^2 (1 - M_j^2) M_i$ , because, from LRT

$$\chi_{jj} = \beta (1 - M_j^2)$$

In the case of  $m$ -component vector spins [195-196] equation (1.45a) generalizes to

$$\underline{h}_i = \sum_j J_{ij} \underline{M}_j - \sum_j J_{ij}^2 \chi_{jj} \underline{M}_i \quad (1.45b)$$



Nakanishi [197] developed two-spin and three-spin cluster theories of SG. The equation for local magnetization for two-spin cluster was exactly identical with TAP equation because the mathematical approximations made in both are equivalent. However, the result of three-spin cluster theory was an expected generalization of equation (1.45):

$$h_i = \sum_j J_{ij} M_j - \sum_j J_{ij}^2 X_j M_i - \sum_j J_{ij} J_{jk} J_{ki} X_j X_k M_i \quad (1.45c)$$

This subtraction of the reaction field is very crucial for SG as shown by Cyrot [198] because the magnitudes of the three fields - Weiss field, Onsager field, and the cavity field are of comparable magnitude in SG. In fact the reaction field can be larger than cavity field in these systems. The physical importance of Onsager field was, of course, realized much earlier when Brout and Thomas [199] showed that the apparent contradiction and violation of fluctuation theorem in naive MFT can be removed by Onsager's method.

Next we come to the crucial evaluation of TAP model - what have been gained by this model? Do they really cure the diseases of the SK model? TAP solution is a saddle point of  $F$  and not a minimum [11]. On the other hand very recently Plefka [200] has shown that the entropy in the TAP model is indeed non-negative. He also obtained an upper bound for the entropy. So far as the crucial question of the occurrence of the phase transition is concerned TAP asserts sharp phase transition like SK model. Plefka [201] obtained the following

lower bound for EA order parameter  $Q$

$$Q > \begin{cases} 1 - \frac{3}{2} t^2 & \text{for } 0 \leq t \leq 2/3 \\ 1 - t & \text{for } 2/3 \leq t \end{cases} \quad (1.46)$$

where,

$$t = (\beta J)^{-1} = \frac{T}{T_c}$$

Since  $Q = 0$  is not allowed for  $t < 1$ , SG seems to be a genuine thermodynamic phase distinct from paramagnetic and ferromagnetic phases. Moreover, Plefka's result agrees with TAP's original result:

$$\begin{aligned} Q &= 1 - t \text{ for } t \leq 1 \\ &= 1 - \alpha t^2 \text{ for } t \rightarrow 0 \end{aligned} \quad (1.47)$$

The slight disagreement in the value of  $\alpha$  ( $= 1.665$  in TAP and  $1.5$  in Plefka) affects the TAP results by little amounts which fall within the error bars of computer simulation. Also notice that all these apparent successes (even if partial) have been achieved without using replica trick.

TAP condition for convergence of their expansion

$$\tilde{J} (1 - \overline{M}^2) \leq T \quad (1.48)$$

was valid only near  $T_g$ . Later Plefka [118] presented a general convergence condition valid for all  $T$ :

$$(\beta \tilde{J})^{-2} > \max \{ (1 - 2Q_2 + Q_4); 2(Q_2 - Q_4) \} \quad (1.49)$$

However recently Owen [202] has corrected some errors in Plefka's calculation [118] and showed that the condition  $(\beta \tilde{J})^{-2} > (1 - 2Q_2 + Q_4)$  is both necessary and sufficient for all temperatures.

But TAP model has its own draw backs. It predicts a cusp in the specific heat exactly similar to the SK model. Besides, in the finite  $Z$  analogue of the TAP model [203] specific heat becomes negative near  $T = 0$  because of negative slope of entropy, for sufficiently large values of  $Z$ , although entropy at  $T = 0$  is zero. Wohlfarth [204] argues that since

$$H\left(\frac{\partial X}{\partial T}\right)_H = \left(\frac{\partial M}{\partial T}\right)_H = \left(\frac{\partial S}{\partial H}\right)_T \quad (1.50)$$

slopes of  $M$ - $T$  and  $X$ - $T$  curves must approach zero as  $T \rightarrow 0$  if third law of thermodynamics is to be obeyed, because the latter claims that "as the temperature tends to zero, the magnitude of the entropy change in any reversible process tends to zero" [205]. TAP clearly violates this law. Wohlfarth accuses the relation (1.47) as the possible cause of this anomaly. So far as the stability of TAP solutions are concerned Bray and Moore [206] computed the eigenvalues of the Hessian matrix  $\frac{\partial^2 (\beta F)}{\partial M_i \partial M_j}$  for each solution of the TAP equation. For stability all the eigenvalues of the Hessian matrix must be positive definite. Though, all the eigenvalues, indeed, turned out to be positive, the eigenvalue spectrum extended down to zero for all  $T \leq T_g$  implying that the solution is always on the limit of stability.

Let us now compare and contrast the SK solution and the TAP solution of the infinite ranged Ising SG model.

Sommers [207], using a diagrammatic expansion of the free energy, summing the relevant diagrams and averaging over the ring diagrams and tree diagrams at successive stages, showed that

- (i) the free energy resulting from averaging over only the ring diagrams is identical with the TAP free energy,
- (ii) averaging over both ring as well as tree diagrams indicates the existence of two solutions one of which is the SK solution. The other solution does not suffer from the unphysical properties of the SK solution.

Sommers' theory predicts that the entropy vanishes as

$$\exp \left[ -2 \left( \frac{2}{\pi} \right)^{1/2} \tilde{J}/T \right] \text{ as } T \rightarrow 0 \quad (1.51)$$

in marked contrast to  $S \propto T^2$  as  $T \rightarrow 0$  as shown by TAP and confirmed by numerical experiments. Bray and Moore [165], in addition to the above point, raised a serious objection against Sommer's solution: "an exponential decay suggests a unique ground state separated by a gap from the first excited state, whereas one expects on physical grounds a quasi-continuum of nearly degenerate ground states".

De Dominicis [208] from a diagrammatic expansion of Martin-Siggale-Rose type, showed how to recover SK and Sommers' solutions in two special cases. Garel [209] pointed out the analogy between the properties of the infinite range Ising

SG and the spinodal behaviour of a ferromagnet. In case of a ferromagnetic Ising model, the system can jump from the unstable branch (SS') to the stable branch (See Figure 13) whereas in the case of an Ising SG "the system is always on the verge of 'jumping' but that there is no thermodynamically stable state onto which the system can jump because of frustration. The system therefore chooses the lowest available energy state, neither stable, nor unstable, that is a typical spinodal situation". Plotting  $Q$  against its conjugate field  $H'$  (see Figure 14) we see that the curve is similar to the  $M$  vs  $H$  curve in Figure 13 except for the fact that the system does not get stable branch to jump on in the former.  $Q < Q_0$  are not allowed as was originally shown by TAP [194]. SK solution falls in this forbidden region.

From the simple consideration of the temperature dependence of the order parameters

$$Q_{\text{TAP}} = \frac{1}{N} \sum_i M_i^2$$

$$Q_{\text{ta}} = \frac{1}{N} \lim_{t \rightarrow \infty} \sum_i S_i(\tau) S_i(t + \tau)$$

and  $Q = [\langle S_i^2 \rangle]_{\text{av}}$ , Young showed [210]

- (a) the SK model is non ergodic,
- (b) a time average gives the same result as a single solution of the TAP equation,

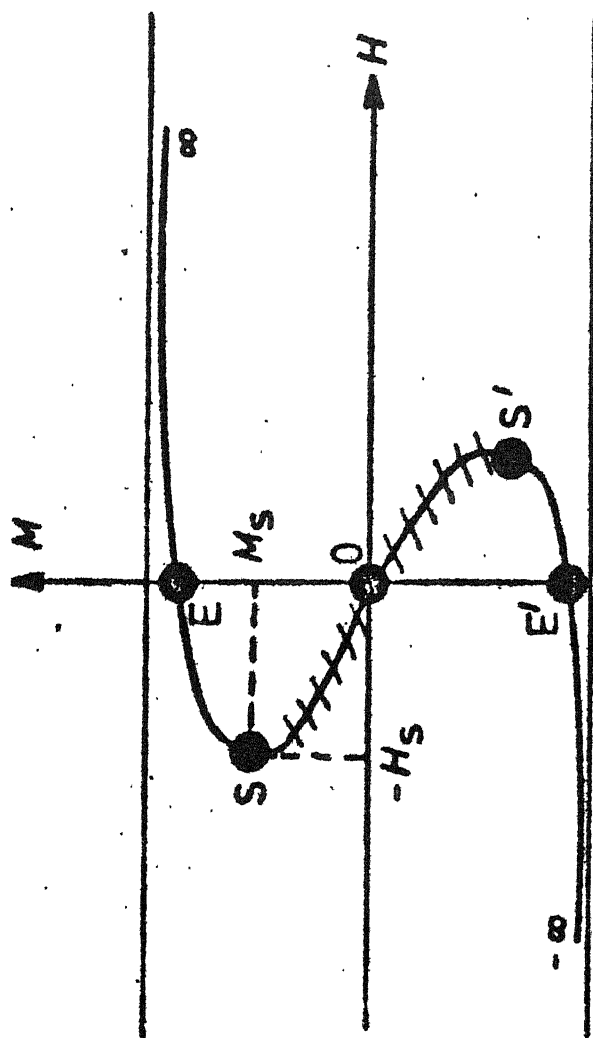


Figure 13 : The ferromagnetic Ising model in a magnetic field ( $T < T_c$ ). The branch  $SS'$  is thermodynamically unstable. (after ref. [209]).

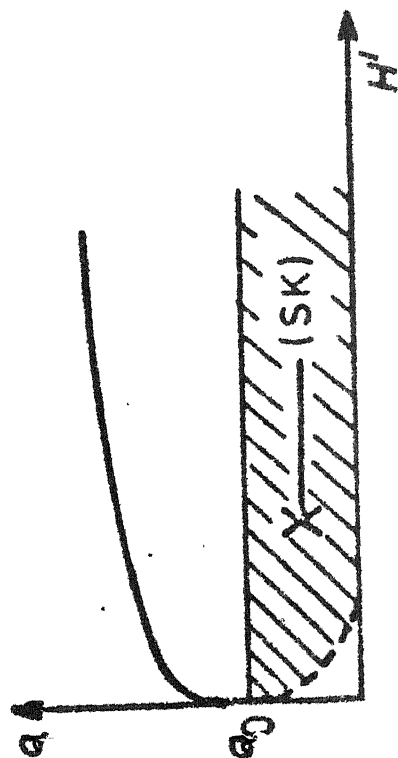


Figure 14 : The TAP solution as a spinodal. The region  $a < a_0$  is not accessible.  $H'$  is the field conjugate to  $a$  (after ref. [209]).

- (c) "ensemble averages correspond to averages over many TAP solutions", and hence, "interference between different solutions causes the ensemble average to decrease more rapidly with T than the results of one solution".

In short, the non-ergodic behaviour arises because of the splitting of phase space into many separate configurations. This behaviour is not a characteristic of only spin glasses but of any glass transition e.g. liquid-glass transition. Jäckle [211] has compared this ergodic-to-nonergodic transition for glasses with that of SG.

Another similar and related question is: Are the famous Maxwell's thermodynamic relations violated in SG? This is a very controversial question. Fogle et.al.'s [212] calorimetric measurements apparently indicated such a failure. They calculated susceptibility from their measured values of  $C(H, T)$  using Maxwell's relation

$$\left(\frac{\partial^2 M}{\partial T^2}\right)_H = \frac{1}{T} \left(-\frac{\partial C}{\partial H}\right)_T \quad (1.52)$$

The calculated values of susceptibility did not agree with the experimentally measured values. But a deeper analysis [213-216] revealed that their data were consistent with Maxwell's relations. On the other hand, from an extensive numerical calculation Soukoulis et.al. [217, 218] concluded that Maxwell's relations were violated in all cases except for the field cooled samples. Of course, the question



remains unsettled.

Kaneyoshi [219] introduced a new method of evaluating the SG transition temperature  $T_g$  beyond its mean field value by using the method of differential operator and carrying out a new type of expansion for the order parameter  $q$  near  $T_g$ .

A full fledged cluster model of SG was developed by Levin, Soukoulis and Grest (LSG)[ 220-222]. The Hamiltonian was divided into inter-cluster and intracluster parts as follows

$$\mathcal{H} = - \sum_{\nu < \lambda} J_{\nu\lambda} S_{\nu} \cdot S_{\lambda} + \sum_{\nu} \sum_{i < j} J_{ij}^0 S_{i\nu} \cdot S_{j\nu} - g\mu_{\beta} \sum_{\nu} \sum_i \underline{H} \cdot \underline{S}_{i\nu} \quad (1.53)$$

where  $S_{i\nu}$  represents the spin the  $i$ th site in the  $\nu$ th cluster.  $J_{ij}^0$  and  $J_{\nu\lambda}$  are the intra cluster and inter cluster exchange interactions respectively.

A very remarkable fact was that the moment per cluster turned out to be temperature dependent which in turn implied a temperature dependent size of the clusters. An equivalent model will be treated later <sup>in</sup> this thesis where the volume of each cluster will be temperature dependent. LSG claimed that this model can explain almost all the experimental results - susceptibility, sp. heat, resistivity and neutron scattering experiment. However, the genesis of these clusters, which has to be universal in order to explain universal properties of all SG, is not convincingly clear. Besides Murani [223]

criticized Soukoulis et.al.'s works on neutron scattering. Soukoulis et.al. subtracted a suitably chosen static component from the total scattering cross section; the static component decreasing monotonically from its maximum value at  $T = 0$  to zero at  $T = T_g$ . The residual (i.e., dynamic) part exhibits peak at unique  $T_g$  independent of  $q$ . However, Murani argued that "the resulting  $q$ -independent peaks in the dynamic component are 'artificially' created additional anomalies in as much as they are assumed to be there to begin with". Such peaks can be obtained even if there were no peaks originally in the total cross section as shown in Figure 15. Moreover, the sharp change of slope at  $T_g$  in the total scattering cross section calculated by Soukoulis et.al. has never been observed experimentally. In a reply to this criticism Levin et.al. [223] attributed the latter discrepancy to their random phase approximation and not the physics of their theory. But their answer to other objections of Murani seem to be less convincing to us.

It is interesting to see that Morandi and Corbelli [224] argued that "no transition with a continuously vanishing order parameter is possible for any physical value of the concentration without violating some necessary conditions on the structure of the internal field distribution function". They attributed the incorrect conclusions of theories of second kind to neglecting the spatial fluctuations. However, from a detailed study of the local field distribution, and,

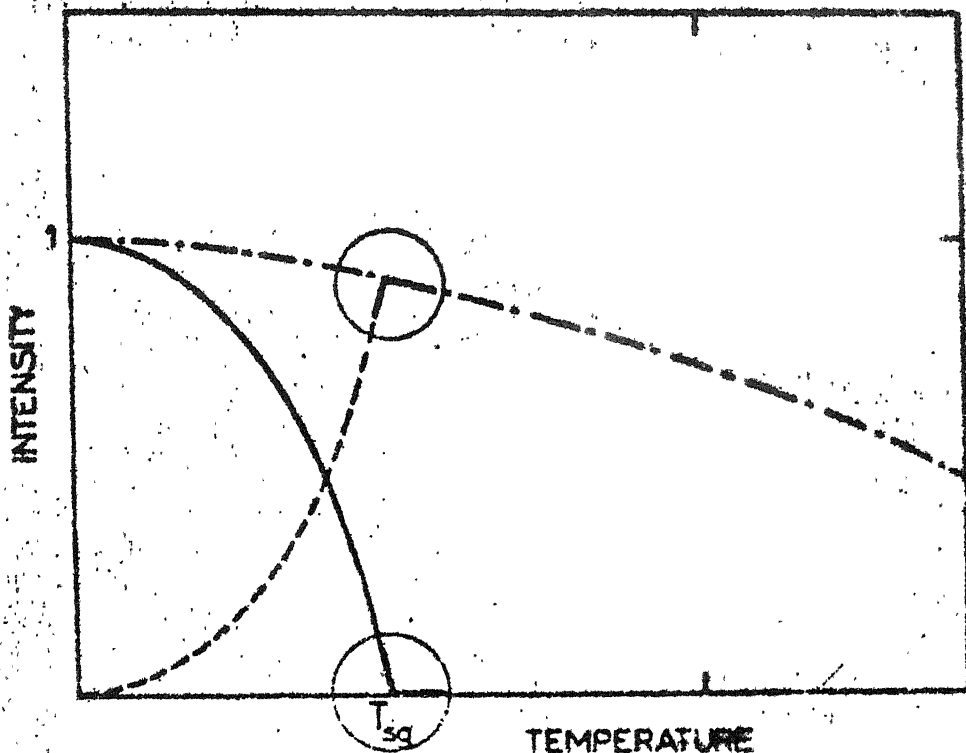


Figure 15 : Schematic variation of the intensity with temperature for one  $q$  value. The chain curve shows a possible monotonic form for the total measured intensity (note no sharp peak originally). The solid curve represents the static (or Bragg) contribution with its sharp discontinuity at  $T_{SG}$  which is circled. The resultant dynamic component is given by the dashed curve together with the chain curve (above  $T_{SG}$ ). It is clear that the sharp peak formed at the junction of the two curves (circled) is the direct results of the assumed sharp discontinuity at  $T_{SG}$  (also circled) in the static component. If  $T_{SG}$  is assumed to be the same for all  $q$  values, it is evident that the dynamic component will have sharp maxima at the same temperature  $T_{SG}$  for all  $q$  (in addition to any other sharp features present in the total intensity curves). (after ref. [223]).

hence, the upper and lower bounds of the transition temperature Corbelli and Morandi [ 225] concluded that a "weak first order transition" takes place at a temperature

$$T_g \propto c^{1/2}, \quad (1.54)$$

because  $Q(T)$  vanishes discontinuously at  $T_g$ .

#### Step V:

Several attempts to understand the critical behaviour of the SG transition have been made using the conventional techniques only with partial success. Most of these results were mutually contradictory and, hence, inconclusive. Since in this review we are mainly concerned with only the MFT we shall not discuss these attempts. The best references are the review articles [ 8],[11], [18]. The upper critical dimensionality seems to be six whereas the lower critical dimensionality four, though these points remain highly controversial. One of the interesting results was obtained recently [ 226 ] applying the ideas of hierarchical Ising model. It was observed that the renormalization group (RG) trajectory is chaotic. By chaotic RG trajectory one means that "as successively longer length scales are viewed using the RG, strong and weak correlations are encountered in a chaotic sequence". The other interesting result is that the upper critical dimensionality is eight for  $m$ -vector SG in the limit  $m \rightarrow \infty$  [227] . Besides Morgan-Pond [ 228] has ,

recently, shown that the size of the critical region for long range (but not infinite) Ising spin glasses is proportional to  $1/Z$  where  $Z$  is the number of interacting neighbours. When compared with phase transitions in other magnetic systems this result is unusual.

### 1.5 Frustration in Spin Glasses

Generally, frustration arises in SG due to the contradiction of the interactions. In canonical SG, for example, due to the oscillating nature of the RKKY interaction and random separation between different spins it may so happen that the information reaching at an arbitrary site- $i$ - from other sites are in conflict with each other leading to frustration of the  $i$ th spin. Since spins connected by frustrated bond do not satisfy the lowest energy (ground state) configuration, the energy of the system is always higher than the corresponding unfrustrated system. It will be shown in this article that frustration distinguishes nontrivial disorder from trivial disorder. Let us consider spins on a square lattice with nearest neighbour exchange interaction with  $|J| = 1$ , as shown in Figure 16, where  $+$  stands for ferromagnetic and  $-$  for antiferromagnetic bond. Notice that the plaquette in Figure 16a is unfrustrated whereas that in Figure 16b is frustrated because all the bonds cannot be satisfied simultaneously in the latter. The number of antiferromagnetic bonds in Figure 16a is even but odd in Figure 16b.

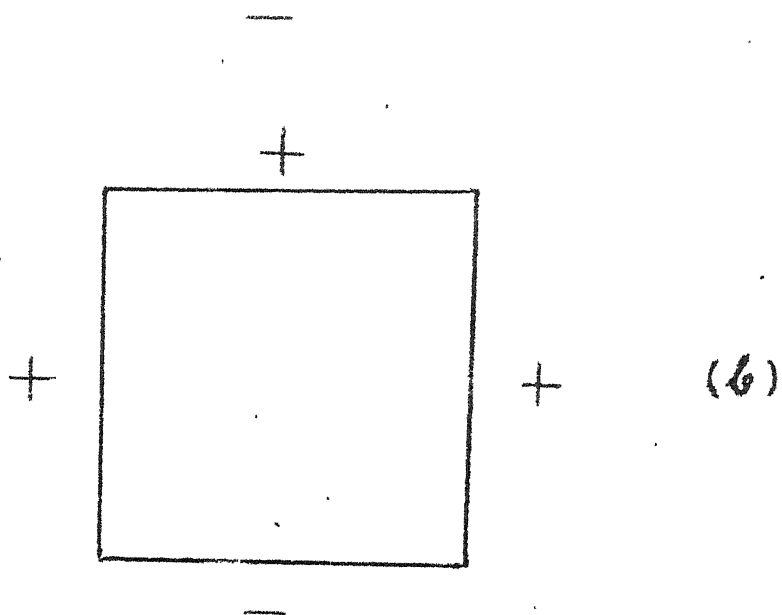
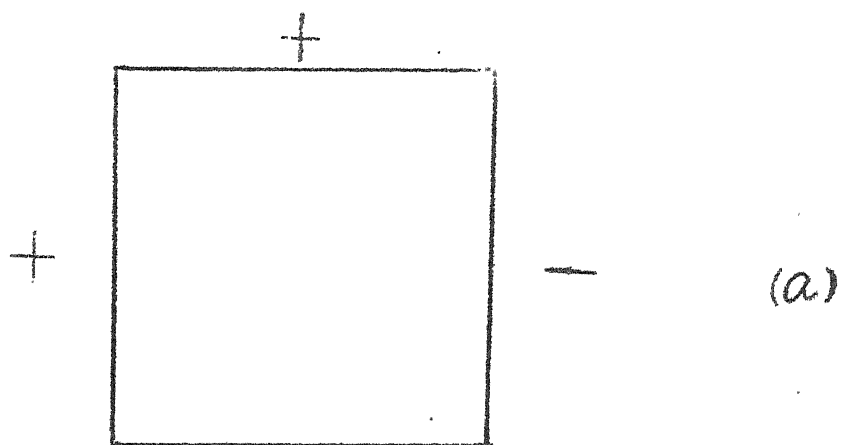


Figure 16 (a) Unfrustrated Square Lattice  
 (b) Frustrated Square Lattice

In a classic paper [229], Toulouse utilized this fact to define frustration function  $\bar{\phi}$  over any closed contour (C) of connected bonds:

$$\bar{\phi} = \prod_{(C)} \text{sign } J_{ij} \quad (1.55)$$

A plaquette is frustrated or unfrustrated depending on whether  $\bar{\phi} = -1$  or  $+1$ . Bethe Lattice does not contain any closed ring and hence totally unfrustrated.

An interesting model of disordered spin system that apparently looked like a SG was introduced by Mattis [230]. This model is defined by

$$H = \sum_{ij} J_{ij} S_i S_j$$

where disorder is introduced via

$$J_{ij} = J \epsilon_i \epsilon_j$$

$\epsilon_i = \pm 1$  randomly on sites. However changing the variables  $S_i \rightarrow S'_i = \epsilon_i S_i$  we recover an ordered problem. In other words, Mattis model does not contain frustration and hence a model of trivial disorder. However, the disorder in Mattis model for quantum spins is nontrivial because the transformation  $S'_i = \epsilon_i S_i$  is not necessarily canonical [231-233].

A very important consequence of frustration is the degeneracy of the ground state. This can be understood geometrically as follows:

"a frustrated bond belongs to two plaquettes (in two dimensional models) and such bonds form a set of strings linking the frustrated plaquettes. The strings are constrained to go through the plaquette centres, and the total length of these strings must be minimal to obtain the lowest energy configuration. There are in general many such minimal pairings, their number is the ground state degeneracy" [234].

Perhaps, the most important impact of the frustration model is that it showed a close relation between quantum field theory and condensed matter physics (and possibly with many other branches of human knowledge) [235-242] proving once again the unity of physics. Psychologists have studied the stable configuration of a group of human beings in the presence of attractions and repulsions between them [234]. The geometry in a SG has similarities with the "parallel transport" of a tangent vector in a curved surface. The misfit between two different lines of transport is expressed by the 'curvature' and 'frustration' in the two cases. In this sense, a frustrated plaquette is 'curved' whereas unfrustrated plaquette is 'flat'. The Mattis model is flat.

The advantage of applying techniques of gauge theory to SG is that frustration can be expressed as a monopole [243]. Rammal and Souletie [15] calls frustration "the third concept in the physics of disorder" - the other two being the concepts of 'percolation' and 'localization'. Percolation concept is associated with the 'dilution disorder' whereas 'localization' describes 'spectral disorder'. In the same spirit we can say



that frustration is associated with "communication disorder" among the various spins.

## 1.6 Dynamical Theories of First Kind:

There are two aspects of spin dynamics in a SG:

- (a) Very Low Temperature ( $T \ll T_g$ ) or short time behaviour and
  - (b) Low Temperature ( $T \lesssim T_g$ ) or long time behaviour.
- Very Low Temperature ( $T \ll T_g$ ) or short time behaviour:

At low temperatures the system gets "trapped" in one of its low lying metastable states, at least for a short time. Consequently, the corresponding excitations, commonly known as spin waves, reflect characteristics of that particular metastable state. In other words, these spin wave excitations arise as small amplitude collective oscillations of the system about the given metastable state when the system responds to an external perturbation. Such excitations have been studied analytically [244-253] as well as numerically [254-261].

Halperin and Saslow [244](HS) developed a hydrodynamic theory of spin waves in SG. The fundamental assumption of the hydrodynamic theory is that the state of the system with small amplitude, long wavelength fluctuation from equilibrium is essentially determined by the long-wavelength variations of the various relevant quantities, e.g., magnetization density, energy density, etc. In that sense HS theory of spin waves is not a microscopic description. The important results of HS

treatment are: linear dispersion law, i.e.  $\omega \propto k$  and that damping is proportional to  $k^2$ . One must remember that these results hold only for  $k \rightarrow 0$  as the latter is the underlying assumption of the hydrodynamic approximation. Andreev's [245] treatment, in the limit of negligibly small external field and relativistic interactions reduce to the HS treatment. These linear HS modes were also obtained by Dzyaloshinskii and Volovik [240]. Saslow [246] extended HS hydrodynamic theory to include remanence and anisotropy effects. Saslow assumed that the external field  $\underline{H}$  is applied along the same direction as the cooling field  $\underline{H}_c$  (i.e. the direction of remanent magnetization  $\underline{M}_r$ ). Later Henley et.al. [247] removed this restriction and took into account a finite angle between  $\underline{H}$  and  $\underline{H}_c$ . In the HS theory two macroscopic variables describing the dynamics were the magnetization  $\underline{m}$  and infinitesimal angle  $\underline{\theta}$  by which the spins are rotated from equilibrium. Saslow [248] introduced an orthonormal triad of unit vectors  $(\hat{n}, \hat{p}, \hat{q})$ , called the spin triad in terms of which individual spin orientations could be specified. The equilibrium orientation for  $(\hat{n}, \hat{p}, \hat{q})$  is called the anisotropy triad  $(\hat{N}, \hat{P}, \hat{Q})$  [248]. Recently Saslow [249] has shown that in the presence of strong uniaxial anisotropy there exist two ground state solutions for  $\hat{n}$  (i.e., the direction corresponding to the cooling field and hence remanent magnetization) - one in which  $\hat{n}$  lies in the  $H - H_c$  plane, called the planar solution, and in the other  $(\hat{n}, \hat{p}, \hat{q})$  is rotated from  $(\hat{N}, \hat{P}, \hat{Q})$  by  $180^\circ$  about the axis midway between  $\underline{H}$  and  $\underline{H}_c$ , called the

$\pi$  -rotation state. The state considered by Henley et.al. [247] was identified with the planar solution.

So far as the macroscopic description of SG are concerned, the latter has similarities with the description of amorphous solids by the theory of elasticity. The spatial displacement vector, velocity and deformation in the elasticity theory are the analogues of the rotation of all spins through the same angle, the temporal derivative and the spatial derivative of the rotation angle, respectively, for SG. However, one remarkable difference is that the different displacements commute whereas different rotations, in general, do not commute.

Takayama [250] gave a microscopic analysis of spin waves in SG using the standard technique of Holstein-Primakoff transformation on the same line as Sherrington's [251] treatment of spin waves in Mattis random magnets. The approximations made in this calculation were valid for  $k \ll 1/R_m$  where  $R_m$  is the mean distance between the spins.

The linear dispersion law was in agreement with HS, but the damping rate turned out to be proportional to  $k^4$  and hence much smaller than that predicted by HS. Takayama [250] also predicted a possible energy gap in the excitation spectrum - the gap arises due to the inclusion of magnetic dipole interaction. However, Takayama's approach was criticized by Barnes [252]. One should expect three distinct spin wave branches due to the spontaneous breaking of the  $O(3)$  symmetry whereas Takayama obtained only one. Barnes [252] obtained all the

three branches in the limit  $k \rightarrow 0$  for a three-dimensional isotropic SG with an axial anisotropy energy. The two transverse branches have identical gaps while the longitudinal does not. The effect of an external magnetic field is to split further the two transverse branches. In general, the dispersion is linear for the longitudinal branch. The quadratic dispersion for the two transverse branches reduce to linear dispersion law for vanishingly small anisotropy. The gap between the macroscopic theories of Andreev [245] and HS [244] and the microscopic theory was bridged by Ginzburg [253] whose microscopic equations reduced to the corresponding hydrodynamic ones in the appropriate limit.

Ching, Huber and coworkers [254-257] and Krey [258] carried out extensive numerical calculations on the existence of spin wave excitations in SG and dynamic structure factor. Their main conclusions were that, for low enough  $k$ , the modes were delocalized for all SG, as predicted by the analytical theories with a remarkable contrast between the metallic SG (e.g. CuMn) and the insulating ones (e.g.  $\text{Eu}_x \text{Sr}_{1-x}\text{S}$ ); in the case of the latter the modes are delocalized over nearly the entire band whereas for the former only low-energy modes are delocalized. Similar results were also obtained by Walker and Walstedt [259-260] whose main quantity of interest in this connection were the density of these low-lying excited states for SG with RKKY interaction and the effect, on these spectra, of the damping of RKKY interaction, dipolar anisotropy, cubic crystal field anisotropy and uniform external field. The

value of the stiffness constant for RKKY-type SG was found to vary as  $c^{4/3}$  and about 25 times smaller than the corresponding value for a comparable ferromagnet [261], consistent with the predictions of HS. One of the assumptions of Watstedt [261], namely, that the exchange stiffness energy varies linearly with area was later confirmed by Banavar and Cieplak [262] thereby lending further support in favour of the existence of spin waves in SG.

#### Low Temperature ( $T \lesssim T_g$ ) or Long Time Behaviour

Thermally activated or quantum mechanical tunnelling commonly known as spin relaxation becomes predominant at higher temperatures or long time scale. These phenomena arise due to the flipping of single spins or clusters of spins as a result of which the system 'hops' from one metastable state to another or to the global ground state. For the time being, we shall be concerned only with the relaxation of the order parameters in SG.

The simplest approach is the use of Langevin equation together with a phenomenological Ginzburg-Landau free energy functional—the so-called time dependent Ginzburg-Landau (TDGL) approach. In case of SG there is no universally accepted form of the free energy functional. However, the TAP free energy is believed to be good enough for this purpose. Kumar and Barma [263] first adopted this procedure where the free energy functional near  $T_g$  is given by

$$\bar{\phi}(Q) = \tilde{J} \left[ \frac{1}{2} \epsilon^2 Q^2 + \frac{2}{3} \epsilon Q^3 + \frac{1}{2} Q^4 \right] \quad (1.56)$$

where

$$\epsilon = \frac{T - T_g}{T_g}$$

and  $\tilde{J}$  is the r.m.s. width of an assumed gaussian distribution of random exchange integrals. The important point to note is that both odd and even powers of  $q$  appear whereas for ferromagnets, from symmetry considerations, only even powers of the order parameter  $M$  appear. This is because dimensionally  $Q$  is equivalent to  $M^2$ . For temperature less than, but very close to,  $T_g$  Kumar and Barma observed power law decay:

$$Q \sim t^{-1/2}, \quad (T \simeq T_g)$$

whereas for  $T > T_g$  the decay is exponential:

$$Q \sim \exp(-\epsilon^2 t/\tau), \quad (T > T_g)$$

Kumar and Barma used TAP free energy where  $M = 0$ . Therefore, they could not derive any expression for magnetization relaxation. Later Shastri and Shenoy [264] generalized the free energy expression to include terms involving even powers of  $M$  as well as terms that couple  $M$  with  $Q$ :

$$\bar{\phi} = \bar{\phi}_Q + \bar{\phi}_M + \bar{\phi}_{MQ} + \bar{\phi}_{MH}$$

where  $\bar{\phi}_Q$  is Kumar and Barma's [263] free energy (1.56)

$$\bar{\phi}_M = a M^2/2, \quad \bar{\phi}_{MH} = -MH$$

and

$$\bar{\epsilon}_{MQ} = -b M^2 Q - c M^2 Q^2$$

In another TDGL approach Ma and Rudnick [265] rederived the  $t^{-1/2}$  behaviour of the spin correlation function. Some fundamental errors of the latter treatment were later pointed out by Sherrington [266]. Besides, Ma and Rudnick's model [265] does not contain any frustration in explicit manner. Hence it was not clear how such a model can represent a SG phase. Hertz and Klemm [267] removed this deficiency of such a model considering short ranged random exchange interactions. The  $t^{-1/2}$  tail was recovered also in this model at and very near  $T_g$  for  $T < T_g$ .

Next better approach is the Master equation approach which was first applied to one dimensional Ising model by Glauber [268] and hence popularly known as Glauber model (also called Kinetic Ising Model). Application of Glauber model to SG was first carried out by Kinzel and Fischer [269] in very simple fashion, which yields exponential decay of the self correlation function for  $T > T_g$ , and power law ( $t^{-1/2}$ ) decay at  $T = T_g$ . This method was generalized for any spin  $S$  by Riess et.al. [270] who observed exponential decay both above and below  $T_g$  and hence much faster than the observed power-law or logarithmic decay. They attributed this discrepancy to the assumption of a single common relaxation time for all spins. In reality, different spins can have different relaxation times.

Kinzel and Fischer's [269] theory was extended by Mody and Rangwala [271] to include cluster effects. Bray et.al. [272] used the same Glauber model to study the average autocorrelation function

$$a(t) = \frac{1}{N} \sum_i \langle S_i(0) S_i(t) \rangle$$

and EA order parameter

$$Q(t) = \frac{1}{N} \sum_i \left\langle \left( \frac{1}{t} \int_0^t dt' S_i(t') \right)^2 \right\rangle$$

which are related by the expression

$$Q(t) = \frac{2}{t^2} \int_0^t dt' (t-t') a(t')$$

They derived an equation of the type

$$\left( \tau \frac{d}{dt} + 1 \right) \langle S_i(0) S_i(t) \rangle = \langle S_i(0) S_i(t) \tanh(\beta h_i(0)) S_i(0) \rangle \quad (1.57)$$

and then made the crucial approximation that the same equation holds even without ensemble average. At low temperatures, the autocorrelation function decays logarithmically whereas the EA order parameter  $Q(t)$  tends to zero for long time  $t$  in agreement with Monte-Carlo results. Bray et.al. [272] approach is probably the only one that yields logarithmic decay even in the homogeneous MFA. It is not clear whether it is consequence of the unconventional approximation used. Very recently a novel idea of Sompolinsky and Zippelius [273] led



to a power law decay of the form  $t^{-\nu}$  for the autocorrelations where the exponent  $\nu \neq \frac{1}{2}$  but depends on temperature as

$$\nu(T) = \frac{1}{2} - \frac{1}{\pi} \left(1 - \frac{T}{T_g}\right) + O\left(\left(1 - \frac{T}{T_g}\right)^2\right) \quad (1.58)$$

A closed expression for the generating functional for the dynamic correlation function was derived by Schuster [274] using De Dominicis [17] method based on an extension of Martin-Siggia-Rose approach [275]. A close relation between SG and many other systems exhibiting  $1/f$  noise (e.g. semiconductors, dielectrics etc.) [276,277] has been indicated by Kogan [278]; the kinetic Ising model was argued to be fairly good description of all these systems.

There is a general trend in the literature to explain the slow decay as a result of spin dynamics of the kind discussed in article 1.6b. However Bray and Moore [279] claimed that such slow decay processes can be dealt with the spin dynamics of the kind described in article 1.6a. Besides, Saslow [280] included both diffusive modes (discussed in article 1.6a) and relaxational modes (discussed in article 1.6b) in the same frame work. For  $k \rightarrow 0$ , rather than having two propagating spin waves, he obtained one diffusive mode and one relaxation mode; the latter having a relaxation time  $T_1$ . On the other hand, for  $ck \gg T_1^{-1}$  only propagating spin waves with linear dispersion were obtained. It is now well established fact that relaxation of both  $m$  and  $q$  are non-exponential (slower than exponential; either power law or logarithmic) [76,24,141,281] for all  $T \leq T_g$ . All the simple mean field type approaches

result of thermal fluctuations exactly as in case of Brownian particles suspended in a liquid.

In a system of uniaxial superparamagnetic particles remanent magnetization relaxes as

$$M_r = M_s e^{-t/\tau} \quad (1.59)$$

where  $M_s$  is the full magnetization when the field is switched off and the relaxation time  $\tau$  is given by

$$\frac{1}{\tau} = \frac{1}{\tau_0} e^{-KV/k_B T} \quad (1.60)$$

$\tau_0$  ( $\approx 10^{-9}$  sec.) arises due to the random thermal fluctuations,  $K$  is anisotropy constant and  $V$  is the volume of the particle. The appearance of  $V$  in the exponent is the most crucial factor in this relaxation. For a particular measurement, characterized by a typical measurement time  $\tau_m$ , particles having volume greater than a critical size appear frozen because their relaxation time will be greater than the measurement time  $\tau_m$ . As an example, a spherical iron particle of radius  $115 \text{ \AA}$  possessing only first order crystalline anisotropy will have a relaxation time of  $\frac{1}{10}$  sec. at room temperature. On the other hand, a particle of radius  $150 \text{ \AA}$  under the same circumstances will have a relaxation time  $10^9$  sec., and hence, extremely stable. Usually  $\tau = 10^2$  sec. is taken as a rough criterion for stability and this occurs at a barrier height approximately equal to  $25 k_B T$ . The temperature at which such stabilization

(popularly called blocking) occurs is called the "blocking temperature".

Wohlfarth [283], in a paper entitled "SG exhibit rock magnetism", drew analogy between the blocking of superparamagnetic clusters and SG "freezing", replacing single domain particles of the former theory by non-interacting clusters of spins in the latter. The evidence for the formation of such clusters at temperatures  $T \gg T_g$  comes from the observed deviation of the  $\chi(T)$  curve from the Curie-Weiss behaviour [73]. Qualitatively speaking, blocked clusters do not respond to external magnetic field and hence do not contribute to susceptibility. This explains a sharp drop in  $\chi(T)$  at  $T_g$ . Though Wohlfarth could not show explicitly the physical origin of the anisotropy energy he gave a list of possible sources [286]:

- (i) surface anisotropy
- (ii) magneto-crystalline anisotropy
- (iii) shape anisotropy
- (iv) magnetostrictive strain anisotropy
- (v) other magnetostatic dipole-dipole effects
- (vi) anisotropic exchange effects.

Wohlfarth [287] realized that a distribution of blocking temperature is closer to physical reality. Therefore, if  $f(T)$  is the distribution of blocking temperatures, the susceptibility is given by

$$\chi(T) = \frac{C}{T} \int_0^T f(T') dT' \quad (1.27e)$$

where  $C$  is the Curie constant. Physically, this expression (1.27c) stands for the fact that only those clusters will contribute to the susceptibility whose blocking temperature  $T_B < T$  i.e., those clusters which have not yet 'frozen'. No first principle derivation of  $f(T)$  was known at that time, though it could be determined from the experimental values of  $\chi(T)$  using the relation

$$f(T) = \frac{d}{dT} (\chi T) / \lim_{T \rightarrow \infty} (\chi T) \quad (1.61)$$

Van Duynveldt and Mulder [288] used this relation to calculate  $f(T)$  for CuMn (0.7 at %) and  $\text{CoO} \cdot \text{Al}_2\text{O}_3 \cdot \text{SiO}_2$  (14.3 at % of Co).

In order to explain the existing neutron scattering experiments Murani [289] introduced the concept of distribution of relaxation times  $g(\tau)$  in a similar manner. The expression for  $\chi(\tau)$  turned out to be similar

$$\chi(\tau) = \frac{C'}{T} \int_0^\tau g(\tau') d\tau' \quad (1.27f)$$

where  $C'$  is a different effective Curie constant. Equation (1.27f) expresses the fact that only those clusters whose life time is less than the measurement time contribute to susceptibility.

### 17.1 Role of Time and Length Scales of Measurements in Theories of Third Kind:

Every experiment probes a certain length of a system for a certain interval of time. A table of characteristic time and length scales of some common experiments are given in Table 1.

TABLE 1

Experiment	Characteristic time	Characteristic length
a.c.susceptibility	$10^{-3}$ sec. - $10^{-1}$ sec.	L
Mössbauer	$10^{-9}$ sec. - $10^{-8}$ sec.	$10^{-13}$ cm.
$\mu$ SR	$10^{-11}$ sec. - $10^{-5}$ sec.	$10^{-13}$ cm.
Neutron scattering	$10^{-12}$ sec. - $10^{-11}$ sec.	$1/q$ .
ESR	$10^{-9}$ - $10^{-10}$ sec.	

We know that blocking occurs when  $\tau \geq \tau_m$ . Therefore,  $T_B$  depends on  $\tau_m$  - greater the frequency lower is  $T_B$  for given V. This explains the frequency dependence of the cusp-temperature in the a.c. susceptibility. The frequency dependence of  $T_g$  for not only the canonical SG but also of insulating SG [290] and transition metal-transition metal SG alloys [291] can be explained by this model. Moreover,

$T_g$  (neutron scatt.)  $> T_g$  (Mössbauer)  $> T_g$  (a.c., susceptibility)  
 because  $\tau_m$  (neutron scatt.)  $< \tau_m$  (Mössbauer)  $< \tau_m$  (a.c.  
 susceptibility). On the other hand, in neutron scattering  
 experiments, higher the value of  $q$  smaller is the size of  
 the clusters explored and hence lower is the blocking temper-  
 ature. Besides, Zibold [292] and Hardiman [293] suggested the  
 idea that ferromagnetic clusters are responsible for the  
 different magnitudes of the frequency shifts in type I and  
 type II SG. The consequence of the different time scales of  
 Mössbauer spectroscopy and  $\mu^+$  SR have been discussed by  
 Hartmann-Boutron [294].

Here one must notice that the frequency dependence of  
 $\chi_{a.c.}(T)$  and difference between the transition temperatures  
 from  $\chi_{a.c.}(T)$  and neutron scattering are not only universal  
 features of SG but also for dipole glasses. In latter kinds  
 of systems, e.g.  $(KBr)_{0.50} (KcN)_{0.50}$ , one measures the frequency  
 dependent dielectric constant instead of susceptibility. Such  
 measurements by Rowe et.al. and by Bhattacharya et.al. [295]  
 showed characteristics similar to SG. Hence, these can also  
 be explained by theories of third kind.

One additional feature of the model is that it does  
 not exhibit any sharp anomaly in the specific heat because  
 blocking is a gradual process where the spin degrees of  
 freedom start freezing from temperatures much above  $T_g$  and  
 continue upto very low temperatures.

This model also explains the Vogel-Fulcher law [296]. But one remarkable feature of this derivation was that the clusters had to be temperature dependent. A detailed comparison of the dynamical properties of rock magnetism and spin glass has been carried out recently by Souletie and by Dormann et.al. [297]. Malozemoff and Imry (MI) [298] measured d.c. susceptibility by using SQUID susceptometer with slowest cooling rate possible. The susceptibility peak turned out to be independent of the cooling rate apparently supporting theories of second kind. But such claims have been criticized by Mydosh [33] on the following grounds -

- (i) MI's experiment used static field cooling thereby 'preparing' the sample in an equilibrium state which was retained so long as the cooling field was kept constant,
- (ii) The time scales of a.c. susceptibility measurements is  $10^{-1}$  sec. as compared to  $10^3$  sec. in SQUID magnetization measurement and the frequency dependence of  $M$  and  $\chi$  may be significantly different in these two regions. Recent experiment of Webb et.al. [299] that shows the ferromagnetic resonance field depends on the rate of cooling as is expected from theories of third kind.

The main drawbacks of such models are the following:

- (i) the susceptibility peak is rounded, though quite sharp, unlike the sharp cusp observed experimentally,

- (ii) while the superparamagnetic particles in rocks are indeed very dilute, they may not be so in SG and hence intercluster interaction may not be negligible,
- (iii) if chemical clustering is assumed to be the dominant source of cluster formation it will be inconsistent with the observed scaling laws,
- (iv) the origin of the anisotropy energy is not clear,
- (v) there is no prescription for calculating  $f(T_B)$  from first principles.

One must notice that Wohlfarth and his group were mainly concerned with susceptibility and specific heat (though of course, they derived Vogel-Fulcher law). An equivalent phenomenological model was suggested by Tholence and Tournier [300] much earlier. The latter model was applied for explaining irreversibility and remanence effects in SG. From a study of the remanent magnetization Tholence and Tournier concluded that a gaussian form is a good representation of the distribution of uncompensated moment of "clouds" of spins, where a "cloud" containing  $n$  spins has a moment proportional to  $\sqrt{n}$ . A rigorous derivation of the latter statement will be presented in this thesis.

So far as the genesis of the clusters is concerned, there have been two distinct views - one group led by P.A. Beck views the cluster formation a consequence of compositional short range order (micromagnetism) and hence have temperature-independent size; the other group views the cluster formation



as a dynamic process and hence clusters possess temperature dependent size. The latter category will be discussed in the Chapter 6 of this thesis.

Although Tholence [301] and later other authors have shown that  $T_g$  obeys a Vogel-Fulcher law [8] rather than Arrhenius law, the controversy is not yet closed.

### 1.7.2 Two Level Systems (TLS) and Anderson-Halperin-Varma (AHV) Model:

We have been discussing relaxation of magnetization in SG. But that is not the only<sup>type</sup> of relaxation studied in such systems. The other important relaxation phenomenon is energy relaxation. Berton et.al. [302] emphasized the difference between these two relaxation processes. In their experiment the SG system "acquired some energy by following a "thermodynamic path"  $C(H, t)$ ,  $T(t)$  (characteristic of the history which the sample has gone to reach the experimental conditions)" and the variation of this energy with time at a given fixed temperature  $T$  in zero field was called the energy relaxation. The interpretation of these results and the comparison of these two types of relaxation needs the introduction of the phenomenological theory of SG as TLS [303] first introduced by AHV [304] as an attempt to explain the low-temperature linear specific heat in all glasses including SG. In this TLS picture it becomes obvious that a splitting of the two levels is essential for energy relaxation but not so for remanent magnetization relaxation.

- AHV's idea consisted mainly of the following parts-
- (a) in a disordered solid (disordered structurally or magnetically) certain atoms or groups of atoms can have two available positions (or, 'directions' of spins in SG) which correspond to two minima of the free energy separated by a finite barrier
  - (b) due to disorder the splitting of the two minima  $\Delta$  is a random function whose probability distribution can be assumed to be a constant.

The energy can be written as

$$\begin{aligned}
 E &= \int_0^{\infty} n(\Delta E) f(\Delta E) d(\Delta E) \\
 &= \int_0^{\infty} n(\Delta E) \frac{\Delta E}{1 + e^{\Delta E/k_B T}} d(\Delta E)
 \end{aligned}$$

and hence, the specific heat is given by

$$C \sim \frac{\pi^2}{6} k_B^2 T n(0)$$

i.e. linear in  $T$ . Though this adhoc phenomenological model worked well, the physical origin of the two-levels was not very clear. The physical origin was later explained by Philips [305] for chalcogenide glasses, by Krishnan [306] for met glasses, and by Cyrot [198] for SG. Cyrot's idea involved two basic ingredients:

- (a) that of Onsager's reaction field, and

(b) that in SG the magnitude of the reaction field can be greater than that of the cavity field. In any SG there will be certain finite number of sites where condition (b) will be satisfied. At those sites reaction field will be along the Weiss field because

$$\text{Weiss field} = \text{Cavity field} + \text{Reaction field}$$

Hence, depending on the direction of the cavity field in any Ising SG there can be two energy levels available - one with energy  $-\langle S_i \rangle H_i^{\text{cav}} - \lambda_i$  (spin parallel to the cavity field) and the other with energy  $+\langle S_i \rangle H_i^{\text{cav}} - \lambda_i$  (spin anti-parallel to the cavity field but parallel to the Weiss field), the first being the stable and the second is metastable state,

$$\lambda_i = \sum_j J_{ij}^2 X_j$$

The same argument is not so obvious in case of Heisenberg model. However, Cyrot [198] considered two possibilities that can introduce uniaxial (or, better, unidirectional) anisotropy - inclusion of dipolar coupling and anisotropy of the reaction field itself. In such cases, all but one of the available states will be metastable.

Similarity of low temperature behaviour of specific heat and similarity of the explanation in terms of TLS have often been cited as an evidence for SG as a nonequilibrium state and hence the terminology glass. The elongation of a

rubber as it responds to an externally applied stress is similar in many respects to the magnetization in a SG. Compliance in the former is the analogue of the susceptibility in the latter, and both exhibit Curie-like behaviour much above glass transition temperatures. Besides, the elongation that remains after withdrawing the stress below  $T_g$  in a stress-cooled rubber can be called thermoremanent elongation (T.R.E.) by analogy with Thermoremanent Magnetization (T.R.M.) Even the time dependences of T.R.M. and T.R.E. seem to be identical, at least qualitatively.

Experimental evidence for TLS nature of SG comes not only from the low temperature linear specific heat but also from the simultaneous study of magnetization and energy relaxations in these systems [307]. In order to study the energy relaxation experimentally the sample is connected with a source of heat and the flow of heat from the latter to the former can be regulated and measured. The rate of loss of energy (i.e., energy relaxation rate) is equal to the rate of gain of energy (which can be measured directly) provided the temperature remains unaltered. For magnetization relaxation no splitting of the two levels is necessary whereas for energy relaxation such splitting is essential. McAlister and Freeman [308] measured the total field cooled magnetization  $M_t$  along the direction of cooling field. Then rotating the sample by  $90^\circ$  very quickly again measured the magnetization. The difference between the two values could be explained by the TLS model

Cyrot's work [198] has been extended [309] to understand the origin of the TLS. Apparently, two level picture indicates Ising character of the spins. Why does, then, a Heisenberg SG exhibit apparent Ising-like behaviour? The potential energy of a classical vector spin  $S_i$  can be written as

$$U = - H_{ic}^\alpha S_i^\alpha - \frac{1}{2} \sum_i^{\alpha\beta} S_i^\alpha S_i^\beta$$

where  $H_{ic}$  is the cavity field at the  $i$ th site,

$$\sum_i^{\alpha\beta} = J_{il} \tilde{\chi}_{lj}^{\alpha\beta} J_{ji}$$

$\tilde{\chi}$  being the local susceptibility tensor with a frozen spin  $S_i$ . The local anisotropy arises due to the anisotropy of local susceptibility. Therefore, there exists a local easy axis (chosen as the polar axis) of magnetization at every site "against which the cavity field  $H_{ic}$  will flight". If the anisotropy field  $\gg$  the transverse component  $H_{ic}$  then we shall get the TLS picture-the two-levels being degenerate provided  $H_{ic} = 0$ . An alternative proof for TLS for the XY model was given by Villian [310].

### 1.8 Metastable States:

Once we have understood the physical nature and origin of the metastable states, we shall next like to count them. One obvious method is to use alternate 'warming' and 'cooling' algorithms for a computer generated finite lattice with a particular type (say, short range, DXXX) of interaction.

and count all the metastable states in which the system settles after each 'cooling' process. Such computations have been carried by Das Gupta et.al.[ 311], and both the postulates (a) and (b) of the phenomenological AHV model were confirmed within the limits of computational error.

Analytical methods are also available for counting the number of metastable states. Since the metastability condition is defined by

$$h_i = \sum_j J_{ij} \sigma_j = \lambda_i \sigma_i, \lambda_i \geq 0$$

the average number of local minima (metastable states) is given by

$$\langle g_0 \rangle = \frac{1}{2} \int_0^\infty \prod_i d\lambda_i \text{Tr}_\sigma \left[ \prod_i \delta \left( \sum_j J_{ij} \sigma_j - \lambda_i \sigma_i \right) \right]_{av}$$

where  $\text{Tr}_\sigma$  implies taking trace over all spin configurations. The prefactor  $\frac{1}{2}$  ensures exclusion of the trivial degeneracy that arises due to the simultaneous flipping of all spins. Notice that the factor  $[ ]_{av}$  is the average number of local minima for a given configuration and hence the trace and the integrations are to be carried out as specified. Tanaka and Edwards [ 312] using this method showed  $\langle g_0 \rangle$  to be of the form  $e^{\alpha N}$  ( $\alpha < 1$  for all spin dimensionalities) for the Ising and XY models with gaussian bond distribution,  $N$  being the number of spins in the system. On the other hand, EA [115] showed  $\langle g_0 \rangle = 2^N$  in two dimensions.

Here again, as in the case of free energy calculation, a controversy arises as to the configuration averaging - whether  $\prod_i \delta (\sum_j J_{ij} \sigma_j - \lambda_i \sigma_i)$  should be first configuration-averaged or the configuration averaging should be carried out at the end. The reason will become clear very soon. Since the average number reveals only a gross feature, it is better to study the distribution of the metastable states  $N_s(E)$ . One can calculate both  $\ln [N_s(E)]_{av}$  and  $[\ln N_s(E)]_{av}$  which are the analogues of free energies for annealed and quenched disorder. Bray and Moore carried out these computations for long-ranged SK model [313], short-ranged (nearest neighbour) Ising model [314], vector spins in the SK model [315], m-component TAP model [316], and observed, in all these cases, that there exists a critical energy  $E_c$  above which both

$$[\ln N_s(E)]_{av} \quad \text{and} \quad \ln [N_s(E)]_{av}$$

become identical. This result was interpreted as expressing absence of correlations between the metastable states for  $E \geq E_c$ . "The onset of correlations between metastable states which develops at  $E_c$  may be regarded as a phase transition with energy taking the role usually played by temperature" [314]. Roberts [317] solved the problem in the presence of external magnetic field for SK model at  $T=0$ . Hertz's dynamical theory [162] can now be interpreted as follows: the barrier between any two local minima diverges in the infinite system and hence any finite-time experiment probes only

fluctuations around a particular minimum. In any finite system  $\varepsilon$  (the theory remains stable for  $t \ll \varepsilon^{-1}$ ) depends on the barrier height. Even the infinite system is never in equilibrium although it takes infinity long time to hop from one metastable state to another. However, Kaplan, on the basis of Lyons-Kaplan theorem [318], showed that "in the classical Heisenberg linear chain with arbitrary nearest neighbour interactions and nonzero but otherwise arbitrary next nearest neighbour interactions there are no metastable states" [319]. Therefore, he commented "that large competition plus randomness does not automatically imply, the existence of a large number of metastable states". Kaplan [319] also compared critically the definition of metastable state adopted by earlier workers with his definition and pointed out the errors committed in these papers. Most of the earliest works defined metastability with respect to single spin flip whereas the correct definition would be 'stability with respect to deviations (or, flips) of arbitrary number of spins'. Finally Kaplan concluded that these earlier papers "leave unanswered the question of the existence of large number of low-lying local minima in two and three-dimensional vector-spin model SG". Therefore the validity of Bray and Moore's objections against the exponential decay of entropy in the Sommers model (see article 1.4.2) remains debatable.



### 1.9 SG Transition as Localization Delocalization Problem

Anderson [320] first made an attempt to apply the concepts of metal-insulator transition and mobility edge to the SG transition. We know that a magnetic ordering takes place in ordered systems when  $\chi(T) J_{\max}(q) = 1$ ,  $J_{\max}(q)$  being the largest eigenvalue of the matrix  $\tilde{J}$ . If  $J(q)$  is maximum for  $q = 0$  the ordered phase is ferromagnetic and for  $q \neq 0$  it is antiferromagnetic. One must notice that in such cases the corresponding eigen function is extended throughout the macroscopic system. This situation changes drastically if the matrix  $\tilde{J}$  is random as in case of SG. A random matrix has both localized as well as extended eigenstates. If the eigenfunction corresponding to the largest eigenvalue of  $J$  is localized, as is usually the case, one gets a hypothetical transition temperature-corresponding to a localized eigenfunction. It is these eigenstates which so convincingly mimic the "clusters" of the Ne'el interpretation of SG phenomenology above  $T_{SG}''$  [8]. But since no phase transition can take place in finite systems the hypothetical transition mentioned above is meaningless. The SG transition was associated with the mobility edge and the random frozen pattern corresponds to the first extended eigenstate of the exchange matrix [321]. This result, however, has been contradicted by Sompolinsky [129] who showed that "SG order is not associated with macroscopic condensation of modes, but is characterized by a distribution of magnetizations throughout

the whole spectrum, each of the  $N$  eigenstates acquires a magnetization of  $O(1)$ ". However, very recently Ueno [322] showed that there exist an infinite number of condensed modes in SK model when analysed in the light of TAP approach. Besides, such a treatment led naturally to the Pa-T hypothesis. Bray and Moore [323] recast the same problem in terms of the eigenstates of the susceptibility matrix  $\chi$  rather than those of the exchange matrix  $J$ . In this case the critical temperature is that at which an eigenvalue of  $\chi^{-1}$  first vanishes. The density of states vanished at zero eigenvalue, in contradiction with the result of Hertz et.al. [321]. The most rigorous quantitative formulation of these ideas was proposed by Ginzburg [324]. But here we are not concerned with localization-delocalization problem. What is more interesting for us is the first indication that probably the correlated behaviour of the spins and the cluster picture are two different manifestations of the same phenomenon. Anderson [8] suggested that the situation in SG is more like a percolation theory than a localization theory. Besides Hertz et.al. [321] commented "the near divergence of the susceptibility associated with the localized states finds a natural physical interpretation in terms of spin clusters".

#### 1.10 Computer Simulation:

Some results of computer simulation have already been reviewed in some related contexts earlier in this thesis.

The results of computer simulation by Binder and coworkers have been extensively reviewed in the references [3,21-24] and those by Walker-Walstedt and by Ching-Huber and coworkers have been reviewed in the References [25-27] . So far as question of the nature of the SG transition is concerned computer simulations give inconclusive answer. However, there is a strong indication that a phase transition may take place, if at all, at  $T=0$ . Similar results were obtained by Reed [325] and by Banavar and Cieplak [326,327] . Reed argued that since a SG can be considered as a unit cell and since the critical temperature was observed to fall with the increase of unit-cell size,  $T_g$  should vanish for a SG. Banavar and Cieplak, on the other hand, applied same type of arguments applied by Abrahams et.al. [328] in the study of Anderson localization. This investigation is based on the dependence of the free energy on the boundary conditions. This result can be interpreted in the light of percolation model because at  $T=0$  radius of the sphere of correlation diverges and hence all the spins belong to the infinite cluster. Recent results of Kinzel [329] supports the view that "SG properties can qualitatively be understood in terms of the relaxation of a distribution of independent clusters", like those in the theories of ~~third~~ kind. Palmer and Pond [330] performed zero temperature computer simulation of the SK model in the absence of a field and Bantilan and Palmer [331] extended it to include nonzero external field. The results of these investigations agreed with Parisi's

replica symmetry-broken theory.

Most of the computer simulation work done so far assumed nearest neighbour gaussian distributed random exchange interactions. Only few attempts [332-334] have been made to work with realistic RKKY interactions. Besides, various type of algorithms have also been tried [335,336]. Fähnle [337] has carried out computer simulation of the distribution of magnetic dipole fields - the distribution is approximately Gaussian. Computer simulations suffer from the drawback that they are performed (i) on finite systems, (ii) for a finite time and (iii) usually for classical spins.

#### 1.11 Some Other Properties

In this section we shall discuss briefly some lesser studied properties of SG. Very little is understood about the transport and galvanomagnetic properties of SG. The low temperature  $T^{3/2}$  behaviour of the resistivity in SG was explained by Rivier and Adkins [338] in terms of the scattering of conduction electrons by the diffusive elementary excitations having very long life time. However, Fischer [339], taking into account the Dzyaloshinski-Volovik modes [240], showed that  $\rho(T) = A T^2 - B T^{5/2}$  with positive A and B in disagreement with experiments as well as Rivier and Adkins [338]. The possible reason for disagreement with Rivier and Adkins was pointed out and the observed  $T^{3/2}$  behaviour was attributed to the existence of predominantly ferromagnetic clusters.

In the papers cited above resistivity means only two contributions to electron scattering -

- (i) scattering due to potential fluctuations, and
- (ii) s-d scattering. On the other hand, Lisyanskii and Trush [340] calculated the following contributions to resistivity:
  - (a) electron-electron scattering (relaxation time  $\tau_e$ )
  - (b) electron nonmagnetic atom scattering (relaxation time  $\tau_i$ )
  - (c) electron-magnetic atom scattering (relaxation time  $\tau_s$ )
  - (d) electron-phonon scattering (relaxation time  $\tau_{ph}$ )
 in the limiting cases of hydrodynamic relaxation ( $\tau_e / \tau_i \ll 1$ ) and kinetic relaxation ( $\sqrt{\tau_i / \tau_{ph}} \ll 1$ ). Campbell et.al.

[341] have developed a semiphenomenological model for resistivity of SG. This model utilized the elementary excitations studied by Walker and Walstedt [259].

The Kondo-effect in SG have been studied by Larsen [342] and Fischer [343] using different techniques. The high temperature  $\ln T$  tail of resistivity and giant thermopower [344] were interpreted by Fischer to be "reminiscent of the Kondo effect".

Theory of magnetoresistance was developed by Mookerjee [345] neglecting the effect of spin dynamics.

PART B

HOMOGENEOUS MEAN FIELD THEORY

OF

COOPERATIVE PHASE TRANSITION MODEL

## CHAPTER 2<sup>\*</sup>

### ISING SPIN GLASS WITH RKKY INTERACTION

In this chapter we shall investigate some particular features of canonical SG, e.g. AuFe and CuMn, which will be modelled as a quenched random Ising spin system in three dimensions. The spins, occupying the lattice sites randomly, interact via indirect Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction, as discussed in Chapter 1. Undamped RKKY interaction is effectively infinite-ranged. The interaction  $J(r)$  is non-random and randomness lies in the occupation of the lattice sites. Such infinite range model [143] is more realistic than the infinitely weak infinitely long-ranged Sherrington-Kirkpatrick (SK) model [140].

#### 2.1 Local Field Distribution : Lorentzian or Gaussian?:

As discussed in section 1.4.1 of Chapter 1, local field distribution (LFD) in Ising SG was derived by Marshall [102], Liu [105], Klein and Brout [103] and Adkins and Rivier [127] to be Lorentzian in mean field approximation (MFA), i.e.,

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\* A part of this chapter has been published in J. Phys. F13 (1983) 365 and the other part has been submitted for publication.

$$P(H) = \frac{1}{\pi} \frac{\Delta}{\Delta^2 + (H - H_0)^2} \quad (2.1)$$

with a mean  $H_0$ ; the width  $\Delta$  is proportional to  $c$ , the concentration of the magnetic constituent. On the other hand, Mookerjee [143] and others [145-147] showed that  $P(H)$  is Gaussian given by equation (1.40) which could be recast as

$$P(H) = \frac{1}{\sqrt{\pi} \Delta} \exp[-(H-H_0)^2/\Delta^2] \quad (1.40a)$$

where,

$$\Delta = (2c Q K_1^2)^{1/2}$$

Our aim in this section is to decide whether the LFD is truly Lorentzian or Gaussian and see under what circumstances they agree with each other.

LFD (2.1) reduces to (1.40a) and vice-versa provided

$$\frac{(H - H_0)^2}{\Delta^2} \ll 1 \quad (2.2)$$

In other words, LFD (2.1) and (1.40a) agree with each other when condition (2.2) is satisfied. Our aim in this section, in other words, is to discuss the physical implications of condition (2.2) in some detail. It will be valid in the following situations:

(i) large  $Q$ , (ii) large  $c$ , (iii) large  $K_1^2$  and (iv) small  $(H-H_0)^2$ . All the conditions (i) - (iii) follow from the same condition — that  $\Delta$  should be large.



$Q$  increases monotonously with decreasing temperature (to be shown in the next section) thereby implying indirectly the validity of condition (2.2) at very low temperatures. Situation (ii) should be interpreted with caution. The concentration  $c$  must be much less than the percolation threshold for long range order so that even short range order can be neglected. Situation (iii) will arise when the magnitude of exchange energy is large. This is physically obvious as large  $J$  would imply large free energy per spin as usual for Lorentzian distribution. Condition (iv) implies that the centre of the distribution can be approximated by a Lorentzian. Although conditions (i) - (iv) look very simple, they have never been stated clearly and systematically in the literature.

Inadequacies of just two order parameters  $M$  and  $Q$  to describe the SG 'phase' have been pointed out by Khurana and Hertz [116], by Parisi [119-121] and others [122,129] and hence the approximate nature of (1.40).

Finally, we conclude that LFD in Ising SG is neither Lorentzian nor Gaussian but a much more complicated function given by the infinite series (1.34). However, in the following chapters in this thesis we shall use (1.40) rather than (2.1) because the former is much more physical than the latter, and poses no problems of diverging moments.

## 2.2 Variation of Order Parameters M and Q with Temperature and External Magnetic Field:

Our aim in this section is to find out, for given canonical SG alloys (characterized by given  $T_c$ ,  $T_g$ ,  $p_{eff}$ ), the correct physical self-consistent solutions of the two coupled integral equations (1.41) and (1.42) numerically by an iterative process.

The characteristics of the given alloys were taken from various experiments (Table III of Reference [83]) and are shown in Table 2.

TABLE 2

Alloy	Concentration of magnetic constituent (in at.%)	$T_c$ (in °K)	$T_g$ (in °K)	$p_{eff}$
<u>AuFe</u>	6.6	2.0	25.5	6.0
<u>CuMn</u>	0.7	3.0	8.0	3.0

The best way to calculate  $T_c$  is from the intercept on the T axis of the high temperature susceptibility  $\chi(T)$ .  $p_{eff}$  was also estimated from susceptibility data.

The choice of the initial input values for M and Q is very crucial for ensuring convergence to the correct physical roots, because the transcendental equations (1.41) and (1.42) have infinite number of solutions. However, the correct self-consistent solutions for  $T = 0$  were obtained as follows:

Let,

$$T_c M + \alpha = A$$

and,

$$T_g \sqrt{Q} = \sigma,$$

then,

$$M = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{-x^2/2} \tanh\left(\frac{\sigma x + A}{T}\right) dx$$

$$Q = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{-x^2/2} \tanh^2\left(\frac{\sigma x + A}{T}\right) dx$$

Again, putting  $\sigma x + A = y$ ,

$$M = \frac{1}{\sqrt{2\pi}\sigma} \int_{-\infty}^{\infty} e^{-(y-A)^2/2\sigma^2} \tanh(y/T) dy$$

$$Q = \frac{1}{\sqrt{2\pi}\sigma} \int_{-\infty}^{\infty} e^{-(y-A)^2/2\sigma^2} \tanh^2(y/T) dy$$

Since  $\lim_{x \rightarrow \infty} \tanh(\pm x) = \pm 1$ ,

$$\begin{aligned} M(T=0, H) &= \frac{1}{\sqrt{2\pi}} \left[ \int_0^{\infty} e^{-(y-A)^2/2\sigma^2} dy - \int_0^{\infty} e^{-(y+A)^2/2\sigma^2} dy \right] \\ &= \text{Er}\left(\frac{A}{\sqrt{2}\sigma}\right) = \text{Er}\left(\frac{T_c M + \alpha}{\sqrt{2}Q T_g}\right) \end{aligned} \quad (2.3)$$

where  $\text{Er}(x)$  is the error function, and

$$Q(T=0, H) = \frac{1}{\sqrt{2\pi}\sigma} \int_{-\infty}^{\infty} e^{-(y-A)^2/2\sigma^2} dy = 1 \quad (2.4)$$

Substituting (2.4) into (2.3) we get

$$M(T=0, H) = \text{Er} \left( \frac{T_c M + \alpha}{\sqrt{2} T_g} \right) \quad (2.5)$$

which can be easily solved self-consistently. In case of AuFe (6.6 at.%), for example,  $M(T=0, H=1 \text{ KG}) \approx 0.01$  together with  $Q(T=0, H) = 1$ . These values for  $M(T=0, H)$  and  $Q(T=0, H)$  were used as initial inputs for  $M(T=0.02, H)$  and  $Q(T=0.02, H)$ . In other words, we move up the  $T$  scale, for fixed  $H$ , solving for  $M(T)$  and  $Q(T)$  self-consistently using  $M(T-\delta T)$  and  $Q(T-\delta T)$  as initial inputs. This is based on the assumption that  $M(T)$  and  $Q(T)$  vary smoothly with temperature, i.e.,  $M(T)$  and  $Q(T)$  differ infinitesimally from  $M(T-\delta T)$  and  $Q(T-\delta T)$  respectively for infinitesimally small  $\delta T$ . Therefore, in this procedure jumping from the physical branch to unphysical branch of solutions, during iteration, is very unlikely. Besides, from analytical solutions of (1.41) and (1.42) we know that in the SG phase, for  $H = 0$ ,  $M(T) = 0$  for all  $T$  and  $Q(T) = 0$  for  $T \geq T_g$ . These facts were used as a check of our computer program. This procedure was repeated for various values of  $H$  (from  $H=1 \text{ KG}$  to  $H=16 \text{ KG}$ ), each time beginning with the corresponding  $M(T=0, H)$  and  $Q(T=0, H) = 1$ .

The variation of  $M$  and  $Q$  with temperature  $T$  for two fixed values of  $H$  ( $H=0$  and  $H = 16 \text{ KG}$ ) are shown in Figure 17.

Since  $Q$  is dimensionally 'similar' to  $M^2$  (both  $\sim S^2$ ) we plotted  $Q$  against  $M^2$ . But no general trend emerged.

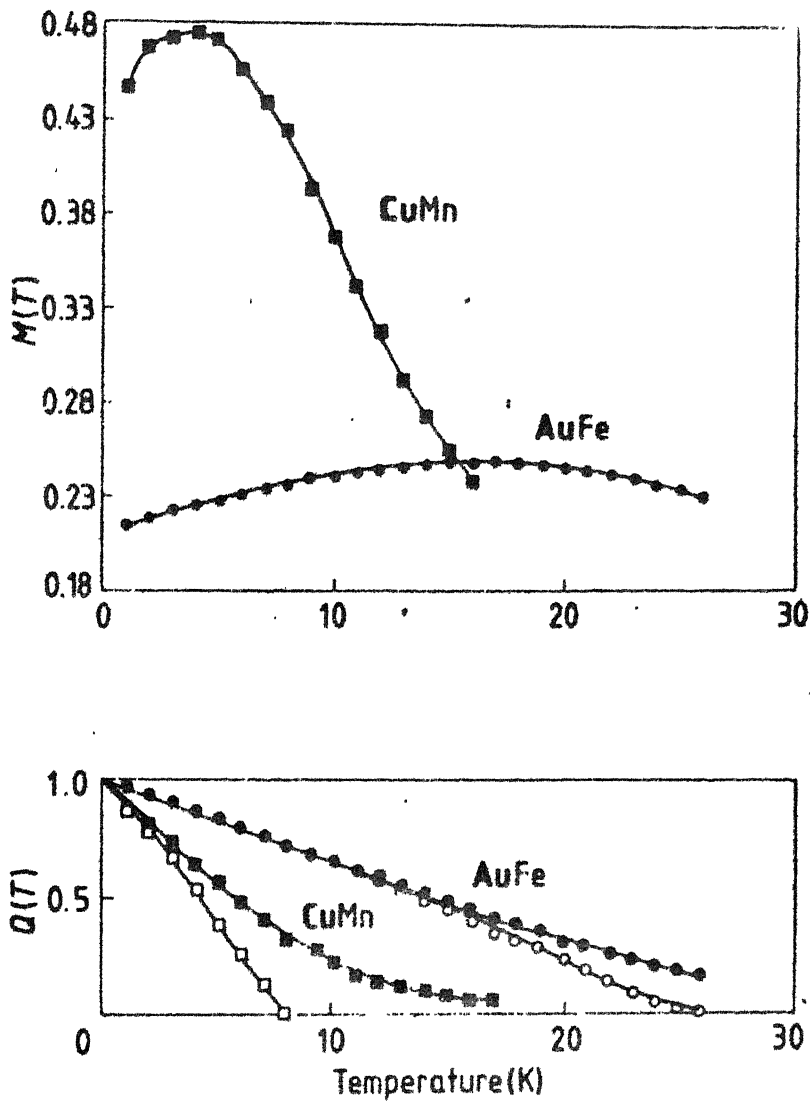


Figure 17 : The temperature variation of the order parameters  $M(T,H)$  and  $Q(T,H)$  without external field (open symbols) and with 16KG applied external field (full symbols) for the two alloys AuFe (1.6 at %) and CuMn (0.7 at %);  $M(T,H = 0)$  is zero throughout.

Surprisingly,  $\Delta Q = Q(H) - Q(O)$ , turned out to be approximately proportional to  $M^2$  over wide ranges of  $M$  and the trend was quite general, as shown in Figure 18 for CuMn (0.7 at.%). This fact will be utilized in Chapter 5.

○ 6K  
● 8K  
□ 10K

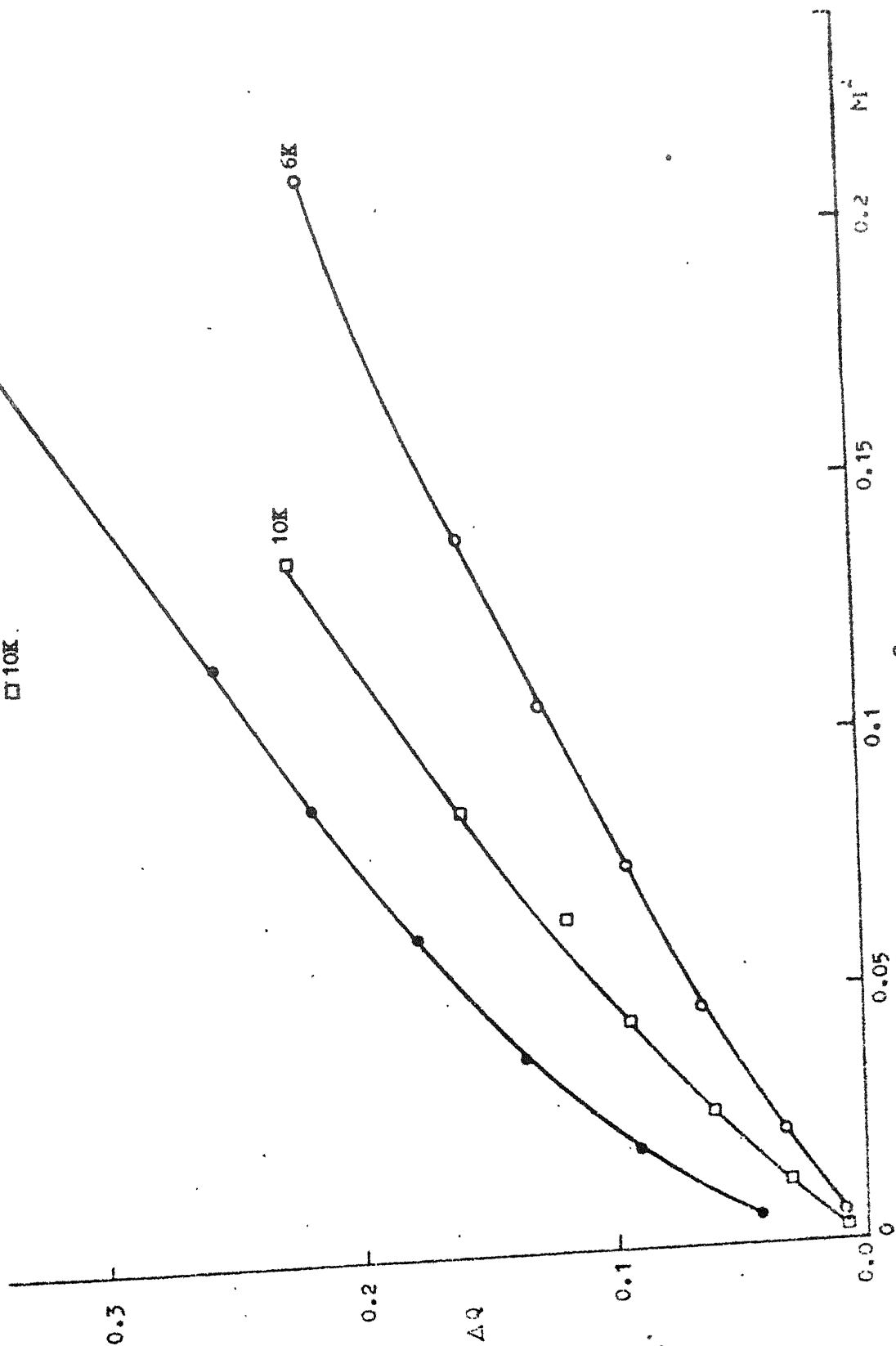


Figure 18 :  $\Delta Q$  versus  $M^2$  for CuMn (0.7 at %) SG at various temperatures.

### CHAPTER 3\*

#### MAGNETORESISTANCE WITHOUT SPIN DYNAMICS : $J_{sd}$

As shown in equation (1.7),  $J_{\text{RKKY}}$  is related to  $J_{sd}$ . Therefore, in order to calculate  $J_{\text{RKKY}}$  from microscopic considerations,  $J_{sd}$  must be known. But in the Heisenberg type hamiltonian for the s-d interaction  $J_{sd}$  is treated as a coarse-grained parameter. It is much more convenient to estimate the latter by comparing experimentally measured quantities (e.g. magnetization, susceptibility, resistivity, magnetoresistance etc.) with the corresponding theoretically calculated values, rather than calculating it exactly from detailed microscopic considerations. In this chapter we shall estimate  $J_{sd}$  in AuFe and CuMn SG by comparing experimental data on magnetoresistance [ 83 ] with the corresponding values calculated using Mookerjee's theory [ 345 ] .

An approximate theory for magnetoresistance of spin glasses was first introduced by Mookerjee [ 345 ] . This theory was formulated using Boltzmann transport equation formalism in spatially random scattering potential. This approach closely followed that of Beal-Monod and Weiner [ 346 ] for dilute alloys.

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\* Contents in this Chapter have been published in J.Phys. F13 (1983), 365.



Let us first clearly systematize the assumptions involved in the theory, as follows [144]:

- (i) there is no short range order (SRO) due to chemical clustering in these alloys,
- (ii) the temperature-concentration range under consideration is far from the Kondo regime, so that the magnetic atoms have well defined local magnetic moments which scatter the conduction electrons,
- (iii) the dominant interaction seen by the conduction electrons are ion-core potential fluctuations  $V = V_A - V_B$  because of alloying and s-d coupling with well defined local magnetic moments at the magnetic sites,
- (iv) the mean free path of conduction electrons  $\delta \gg a$ , the lattice constant, so that we are far from the mobility edge and all conduction electrons are delocalized,
- (v) the resistivity can be calculated using the Boltzmann transport equation which is valid in the limit  $k\delta \gg \pi$ ,
- (vi) Born approximation can be used. The latter approximation requires  $ka \gg \pi$ , which is not necessarily guaranteed by assumptions (iv) and (v).
- (vii) the potential fluctuations and s-d coupling are isotropic and local,

- (viii) the concentration of the magnetic component is small, so that there is no micromagnetic clustering. The dominant order parameter of interest is only the on-site freezing, so that

$$[\langle S_i \rangle \langle S_j \rangle]_{av} = Q(T) \delta_{ij}$$

- (ix) the effect of spin-orbit interaction on magnetoresistance may be neglected,  
 (x) the spin dynamics is ignored so that

$$R(\omega) = \int_{-\infty}^{\infty} [\langle S_i(t) S_i(0) \rangle]_{av} e^{i\omega t} dt$$

may be replaced by  $Q(T) \delta(\omega)$ , i.e., the spin system reaches equilibrium instantaneously. This last assumption will be relaxed in Chapter 5 so as to incorporate the effect of spin dynamics.

Mookerjee [345] showed that

$$\begin{aligned} \Delta\rho = \rho(H) - \rho(0) = & -c R_o J_{sd}^2 [M(H) \tanh(\frac{g\mu_B H}{2k_B T}) \\ & + 2\{Q(H) - Q(0)\} \{1 - \frac{J_{sd}^2}{V^2} S(S+1)\} \\ & - \frac{2 J_{sd}^2}{V^2} M(H) Q(H) \tanh(\frac{g\mu_B H}{2k_B T})] \end{aligned} \quad (3.1)$$

where,

$$R_o = \left(\frac{3\pi m}{4\hbar e^2}\right) \left(\frac{1}{n E_F}\right),$$

$n$  being the number of conduction electrons per unit volume. For AuFe and CuMn the ratios of the s-d coupling to the potential fluctuations, i.e.,  $|J/V|$  are about 0.19 and 0.16 respectively [347], so that we may neglect second and higher order terms in  $J_{sd}/V$  in (3.1). Thus

$$\Delta\rho = -c R_0 J_{sd}^2 \left[ M(H) \tanh\left(\frac{g\mu_B H}{2k_B T}\right) + 2\{Q(H) - Q(0)\} \right] \quad (3.2)$$

$M(H)$  and  $Q(H)$  were calculated as reported in Chapter 2.  $R_0$  was calculated by Nigam and Majumdar [83] assuming that the Fermi energy  $E_F$  and the number of electrons per unit volume  $n$  are the same as those for the corresponding host metals. Hence,  $\Delta\rho/c R_0 J_{sd}^2$  was calculated using (3.2) and shown in Figures 19a and 20a for AuFe (6.6 at %) and CuMn (0.7 at %) alloys respectively. The corresponding experimental values for  $(\Delta\rho/c R_0)$  for the same two alloys are shown in Figures 19b and 20b for comparison.

Let us now discuss the general features of the  $H$  and  $T$  dependence of magnetoresistance in AuFe and CuMn SG. Theory and experiment show qualitative agreement with a quadratic  $H$  dependence for small values of  $H$ , going over to  $H^\alpha$  ( $1 < \alpha < 2$ ) as we go to higher fields. Quantitatively, the agreement is also fairly good. Theoretical curves show the following features for the magnetoresistance isotherms: high-temperature magnetoresistance is small and the isotherms lie near the base line; as the temperature decreases negative magnetoresistance increases and the isotherms lie below the high

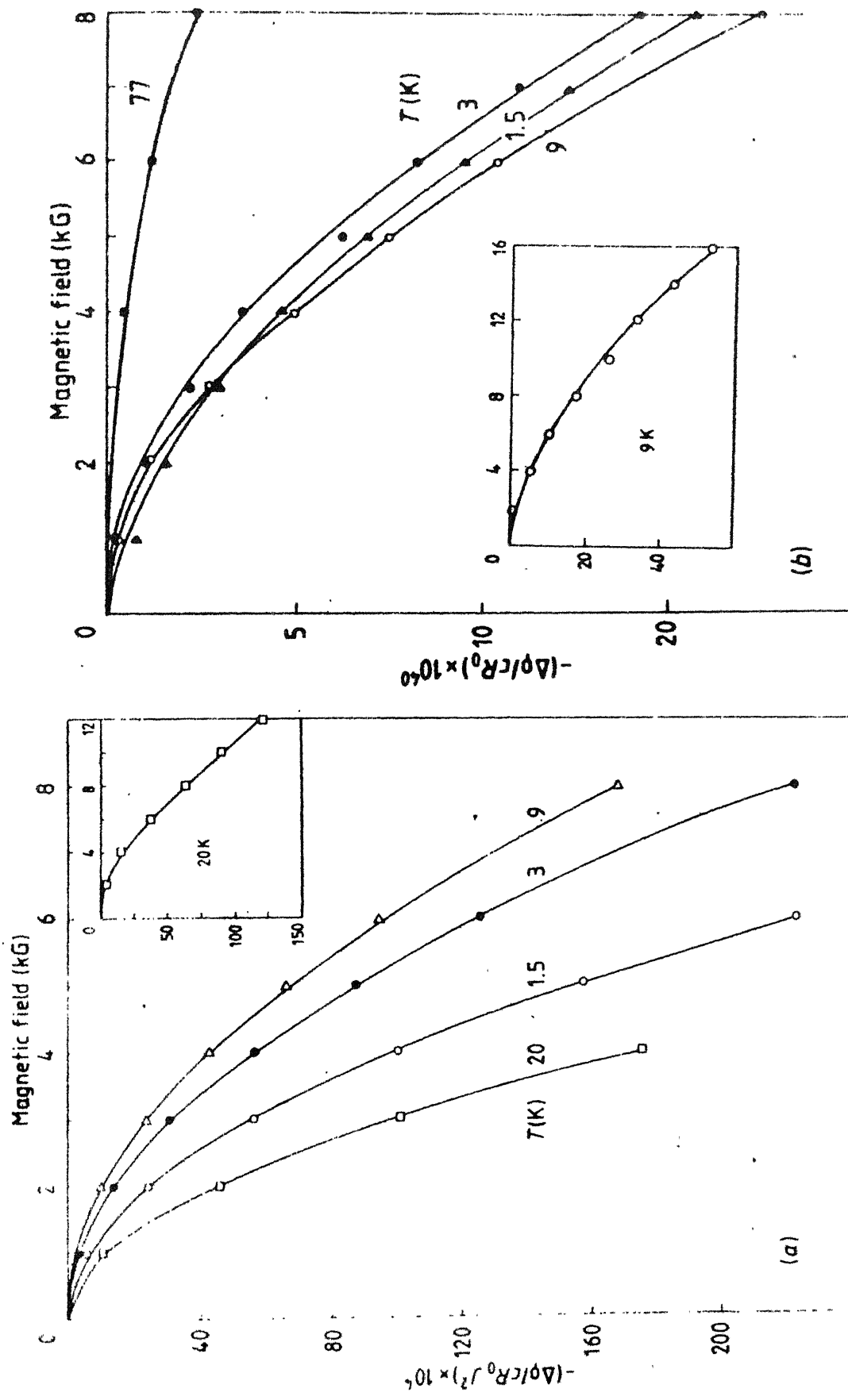


Figure 19 : (a) Theoretical value of  $\Delta P/cR_0 J^2$  for AuFe (6.6 at %) shown as a function of magnetic field for various temperatures. The inset shows a wider variation with field; the abscissa scale is contracted ten-fold with respect to the figure.  
 (b) Experimental  $\Delta P/cR_0$  for AuFe as reported by Nigam and Majumdar (1981) for comparison.

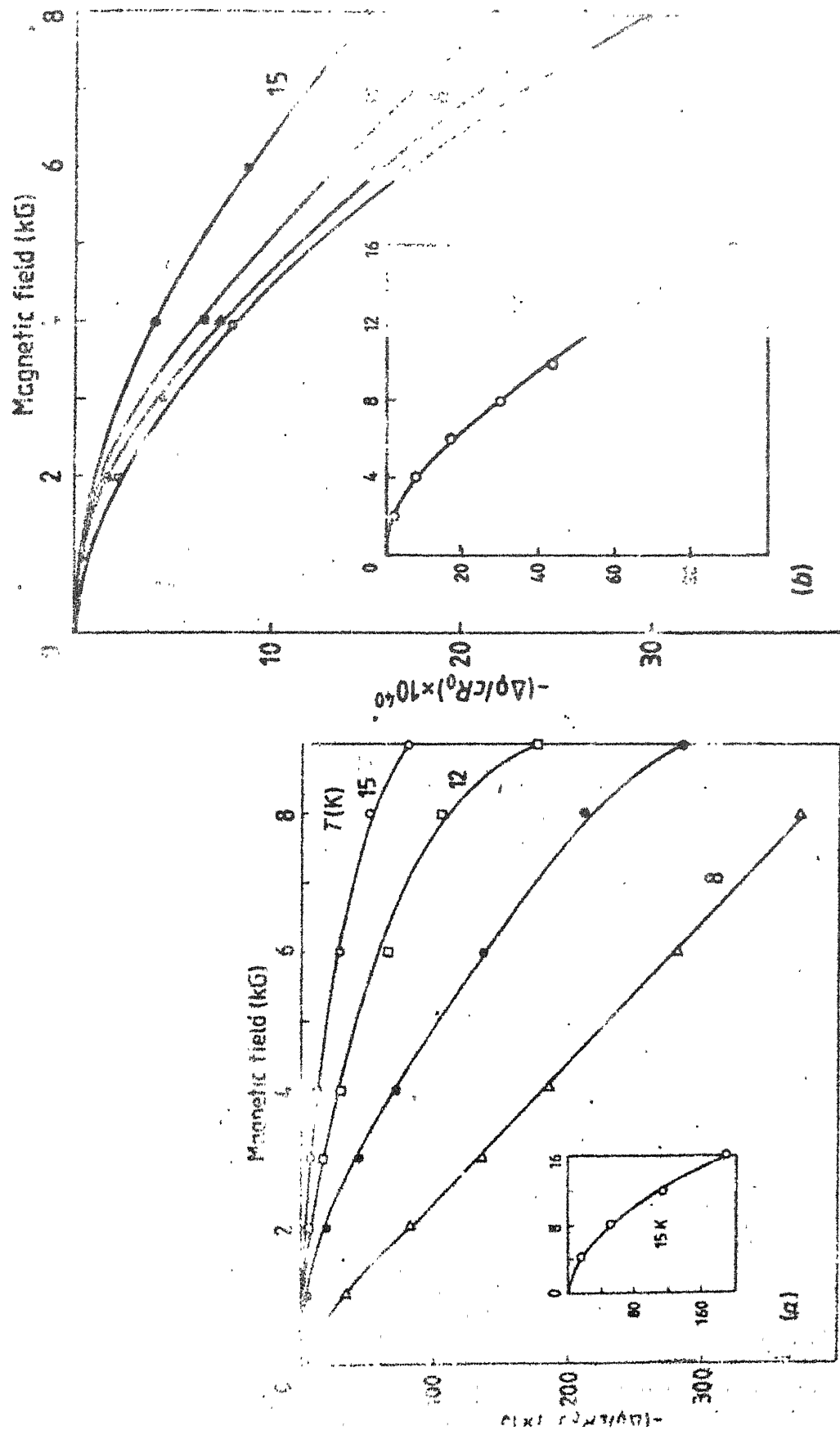


Figure 20 : (a) Theoretical value of  $\Delta\rho/cR_0 J^2$  for CuMn, (0.7 at %) shown as a function of magnetic field for various temperatures. The inset shows a wide range of magnetic field with field; the abscissa scale is contracted ten-fold with respect to the main figure.

(b) Experimental  $\Delta\rho/cR_0$  for CuMn as reported by Majumdar and Majumdar for comparison.

temperature ones. At still lower temperatures magneto-resistance again increases and the isotherms cross those for high-temperatures and lie above them. In other words,  $\Delta\rho$  as a function of  $T$  has a maximum. Since at these temperatures  $\rho(T)$  itself increases with  $T$  but with no sharp features, we also expect  $\Delta\rho/\rho$  to have a maximum. This feature was reported by Nigam and Majumdar [83]. Theoretically, this maximum is around 20K for AuFe and between 8 and 6K for CuMn. Experimentally, this maximum temperature is much lower: below 9K for AuFe and below 4.5K for CuMn. More careful theory and analysis of experimental data are required to obtain quantitative agreement.

Finally,  $J_{sd}$  was estimated from

$$J_{sd} = \left[ \left( \frac{\Delta\rho}{c R_0} \right)_{\text{expt.}} / \left( \frac{\Delta\rho}{c R_0 J_{sd}^2} \right)_{\text{theo.}} \right]^{1/2}$$

where  $\left( \frac{\Delta\rho}{c R_0} \right)_{\text{expt.}}$  were taken from Nigam and Majumdar's [83] experimental data. We present the calculated values of  $J_{sd}$  in Table 3. The variation of  $J_{sd}$  with  $H$  and  $T$  arises due to both the approximations involved in the theory as well as random scatter of the experimental data. Besides, in estimating various parameters that enter the theoretical calculations, experimental data had to be taken from various sources. As a result, one could not estimate and compare the relative accuracy or reliability of different data. Ideally, either there should exist a comprehensive microscopic theory which would enable us to calculate all the parameters

abinitio, or all experiments should have been performed on the same samples, preferably by the same group. In Table 4 the values of  $J_{sd}$  predicted previously by various authors have been shown together with ours.

TABLE 3

<u>Al:Fe</u>	1	2	3	4	5	6	8	10	12	14	16	J	J <sub>sd</sub>
<u>H</u>													
<u>T</u>													
3.0	1.71	1.7	1.65	1.58	1.67	1.6	1.59	1.58	1.58	1.55	1.52	1.61	
9.0	2.02	2.05	2.12	2.13	2.11	2.08	2.02	1.96	1.90	1.85	1.80	2.00	1.52 <sup>+</sup> -0.085
20.0	1.06	0.77	0.90	0.88	-	0.96	0.98	0.99	0.99	0.99	0.99	0.95	
<u>Cu:Mn</u>													
4.2	1.02	1.02	0.97	0.95	-	0.95	0.98	0.98	0.98	0.98	0.99	0.98	
6.0	-	0.60	0.64	0.64	-	0.67	0.70	0.73	0.76	0.78	0.81	0.70	
10.0	0.67	0.66	-	0.64	-	0.66	0.68	0.71	0.73	0.75	0.77	0.70	0.80 <sup>+</sup> -0.041
12.0	-	0.91	-	0.94	-	0.90	0.89	0.89	0.89	0.89	0.90	0.90	
15.0	-	1.15	-	1.08	-	1.07	1.06	1.04	1.03	1.02	1.01	1.06	

H is in KG, T in °K, J in eV



TABLE 4

Reference	Alloy	M	$\chi_c$	C	$\rho$
[ 387 ]	CuIn	2.2±0.15			
[ 78 ]	-do-	3.3			
[ 388 ]	-do-		0.55	0.47	0.29
[ 144 ]	-do-				0.80 ±0.04
[ 347 ]	AuFe			0.56	0.75
[ 144 ]	-do-				1.52 ±0.09

The table gives the predicted values for  $J_{sd}$  from various experimental data for M,  $\chi_c$ , C or  $\rho$ . The values are quoted in eV.

## CHAPTER 4<sup>\*</sup>

### SPIN DYNAMICS AND RESISTIVITY : INADEQUACY OF SINGLE RELAXATION TIME

It is now experimentally well established that there exists a distribution of relaxation times (DRT) in SG [85,288]. On the other hand, all mean field theories eg.[270,271] of cooperative phase transition from paramagnet-to-SG phase, except recent dynamical theories of Sompolinsky [117] and Hertz [162], yield a single relaxation time. Even in the Sompolinsky-Hertz dynamical approach to infinitely weak infinitely long ranged SK model DRT appears only for finite sized systems; in the thermodynamic limit all the relaxation times diverge.

In order to establish inadequacies of single relaxation time we present some further evidences in this chapter. We shall show how single relaxation time is not only inadequate to account for the observed relaxation of order parameters but also insufficient to explain the variation of resistivity with temperature.

Our brief discussion of relaxational spin dynamics will have some new features.

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<sup>\*</sup>Submitted for publication.

#### 4.1 Spin Dynamics

The system under consideration is modelled by a collection of  $n$  spins randomly distributed over  $N$  lattice sites. The spins interact with one another with predominantly indirect RKKY exchange interaction. First let us consider classical Ising spins with a hamiltonian

$$H = - \sum J(|\underline{r}_i - \underline{r}_j|) S_i S_j \quad (4.1)$$

Defining  $M(t) = [\sigma_i(t)]_{av}$  and  $Q(t) = [\sigma_i^2(t)]_{av}$ , as before, where

$$\sigma_i(t) = \sum_{\{S\}} S_i P(\dots, S_i, \dots; t)$$

and writing the usual Master equation for the spin configuration  $P(\dots, S_i, \dots; t)$  we get

$$\left(\frac{\eta}{k_B T}\right) \frac{dM}{dt} = -M(t) + [\langle \tanh(\beta h_i) \rangle]_{av} \quad (4.2)$$

and

$$\left(\frac{\eta}{2k_B T}\right) \frac{dQ}{dt} = -Q(t) + [\sigma_i(t) \langle \tanh(\beta h_i) \rangle]_{av} \quad (4.3)$$

where,

$$h_i = \sum_j J_{ij} S_j$$

Equations (4.2) and (4.3) are of central importance in the discussion of the spin dynamics. We carry out the configuration averages in (4.2) and (4.3) in the Bethe-Pierls-Weiss approximation to get

$$\begin{aligned} \left(\frac{\eta}{k_B T}\right) \frac{dM}{dt} = & - \left(1 - \frac{T_c}{T}\right) M(t) - \frac{1}{3} \left(\frac{T_c}{T}\right)^3 M^3(t) \\ & - \frac{T_c T_g^2}{T^3} M(t) Q(t) + \dots \end{aligned} \quad (4.4)$$

and

$$\left(\frac{\eta}{2k_B T}\right) \frac{dQ}{dt} = - \left(1 - \frac{T_g^2}{T^2}\right) Q(t) + \frac{T_c}{T} M^2(t) - \frac{T_g^2}{T^2} Q^2(t) \quad (4.5)$$

We shall solve equations (4.4) and (4.5) for low concentration (SG regime) alloys at various temperature ranges. For such alloys  $T_c < T_g$ .

(a) High Temperature ( $T > T_g > T_c$ ) Solutions:

We know that the asymptotic values are  $M = 0$  for all  $T$  and  $Q = 0$  for  $T > T_g$ . Hence, if we start with small  $M$  and  $Q$  at  $t = 0$ , they will continue to be small at all times, So, in this regime

$$\left(\frac{\eta}{k_B T}\right) \frac{dM}{dt} = - \left(1 - \frac{T_c}{T}\right) M(t) \quad (4.6)$$

and

$$\left(\frac{\eta}{2k_B T}\right) \frac{dQ}{dt} = - \left(1 - \frac{T_g^2}{T^2}\right) Q(t) + \left(\frac{T_c}{T}\right) M^2(t) \quad (4.7)$$

Solving (4.6) and (4.7) with the boundary conditions  $M = M(0)$  at  $t = 0$  and  $Q = Q(0)$  at  $t = 0$ , we get

$$M(t) = M(0) \exp(-t/\tau_c) \quad (4.8)$$

$$Q(t) = [Q(0) - DM^2(0)] \exp(-2t/\tau_g) \quad (4.8)$$

$$+ DM^2(0) \exp(-2t/\tau_g) \quad (4.9)$$

where,

$$\tau_c^{-1} = \frac{k_B T}{\eta} \left(1 - \frac{T_c}{T}\right) \quad (4.10)$$

$$\tau_{g>}^{-1} = \frac{k_B T}{\eta} \left(1 - \frac{T_g^2}{T^2}\right) \quad (4.11)$$

and

$$D^{-1} = 1 - \frac{T_g^2}{T T_c} \quad (4.12)$$

Such a behaviour of  $M$  and  $Q$  is characteristic of paramagnetic phase. The novel feature of our solutions is the coupled relaxation of  $M(t)$  and  $Q(t)$  as can be seen from equation (4.9).

As  $T \rightarrow T_g$ ,  $Q$  exhibits critical slowing down. However,  $M$  does not show any critical slowing down near  $T_g$ . Physically this is expected, because, as discussed in Chapter 1, only the order parameter susceptibility diverges at  $T_g$  whereas linear susceptibility associated with  $M$  exhibits only a cusp at  $T_g$ .

At temperatures very close to (but not exactly at)  $T_g$ , both the first and third terms in (4.5) cannot be neglected. For convenience, we choose our initial condition as  $M(0) = 0$ . Then (4.5) reduces to

$$\left(\frac{\eta}{2k_B T}\right) \frac{dQ}{dt} = - \left(1 - \frac{T_g^2}{T^2}\right) Q(t) - \frac{T_g^2}{T^2} Q(t)$$

Completing the squares on the right hand side we get,

$$\left(\frac{\eta}{2k_B T}\right) \frac{dQ'}{dt} = - \left(\frac{T_g^2}{T^2}\right) (Q'^2 - b^2)$$

where,

$$Q' = Q + b$$

and

$$b = \frac{1}{2} \left( \frac{T^2}{T_g^2} - 1 \right)$$

Therefore,

$$Q(t) = \frac{2b Q(0) \exp \left[ -\frac{2k_B T}{\eta} \left( 1 - \frac{T_g^2}{T^2} \right) t \right]}{Q(0) \left[ 1 - \exp \left\{ -\frac{2k_B T}{\eta} \left( 1 - \frac{T_g^2}{T^2} \right) t \right\} \right] + 2b}$$

which is still exponential for very large as well as very small  $t$ . At  $T = T_g$

$$\left( \frac{\eta}{2k_B T} \right) \frac{dQ}{dt} = \left( \frac{T_c}{T} \right) M^2(t) - \left( \frac{T_g^2}{T^2} \right) Q^2(t) + \dots$$

For large values of  $t$ ,  $Q(t) \sim t^{-1}$ , since the first term is exponential and dominates only at short times.

#### (b) Low Temperature ( $T < T_g$ ) Solutions:

In this regime  $Q(\infty) \neq 0$  although  $M(\infty)$  is still zero. Let,  $Q(t) = Q(\infty) + q(t)$  and  $M(t) = M(\infty) + m(t)$ , where  $m(t)$  and  $q(t)$  are small amplitude fluctuations around the equilibrium values  $M(\infty)$  and  $Q(\infty)$ , at time  $t$ .

Then from (4.4) we get

$$\left(\frac{\eta}{k_B T}\right) \frac{dm(t)}{dt} = - \left(1 - \frac{T_c}{T}\right) m(t) - \frac{T_c T_g^2}{T^3} m(t) Q(\infty) \quad (4.13)$$

On the other hand, from (4.5) we get

$$Q(\infty) = 0, \text{ or } Q(\infty) = \left(1 - \frac{T^2}{T_g^2}\right) \quad (4.14)$$

It is easy to see that the latter solution is the only stable solution for  $T < T_g$  while the former is the stable solution for  $T > T_g$ . Hence substituting (4.14) into (4.13) and solving it

$$M(t) = M(0) \exp\left[- \left(\frac{k_B T}{\eta}\right) \left(1 - \frac{2T_c}{T} + \frac{T_c T_g^2}{T^3}\right) t\right] \quad (4.15)$$

Similarly, from (4.5) we get

$$\begin{aligned} \left(\frac{\eta}{2k_B T}\right) \frac{dq(t)}{dt} = & - \left(1 - \frac{T_g^2}{T^2}\right) Q(\infty) - \frac{T_g^2}{T^2} Q^2(\infty) \\ & - \left(1 - \frac{T_g^2}{T^2}\right) q(t) + \frac{T_c}{T} m^2(t) - \frac{2T_g^2}{T^2} Q(\infty) q(t) \end{aligned} \quad (4.16)$$

Substituting (4.14) into (4.16) and solving

$$Q(t) = Q(\infty) + \left[ Q(0) - Q(\infty) - \frac{m^2(0)}{\left(2 + \frac{T_g^2}{T T_c} - \frac{T_g^2}{T^2} - \frac{2T}{T_c}\right)} \right] \exp(-2t/\tau_g)$$

$$+ \frac{m^2(0)}{(2 + \frac{T_g^2}{TT_c} - \frac{T_g^2}{T^2} - \frac{2T}{T_c})} \exp\left[-\left(\frac{2k_B T}{\eta}\right)\left(1 - \frac{2T_c}{T} + \frac{T_c T_g^2}{T^3}\right)t\right] \quad (4.17)$$

where,

$$\tau_{g-1}^{-1} = \frac{k_B T}{\eta} \left(\frac{T_g^2}{T^2} - 1\right)$$

It is easy to verify that  $(1 - \frac{2T_c}{T} + \frac{T_c T_g^2}{T^3}) = (1 - \frac{T_c}{T})^2 + \frac{T_c}{T} (\frac{T_g^2}{T^2} - 1) > 0$  for  $T < T_g$ , i.e., for both  $T_c < T < T_g$  and  $T < T_c < T_g$ , and hence  $M(t)$  in equation (4.15) and the last term in (4.17) always decay to zero. Thus, even below  $T_g$  both  $M$  and  $Q$  decay exponentially to their equilibrium values in the homogeneous mean field theory. There is a unique single relaxation time  $\tau_{g-1}$  for  $T < T_g$  which is not true as stated earlier.

Very close to, but below,  $T_g$ , we have

$$\left(\frac{\eta}{2k_B T}\right) \frac{dq}{dt} = -\left(\frac{T_g^2}{T^2} - 1\right) q(t) - \frac{T_g^2}{T^2} q^2(t)$$

and hence

$$Q(t) = Q(\infty) + \frac{2b' \{Q(0) - Q(\infty)\} \exp\left[-\frac{2k_B T}{\eta} \left(\frac{T_g^2}{T^2} - 1\right)t\right]}{\{Q(0) - Q(\infty)\} \{1 - \exp\left[-\frac{2k_B T}{\eta} \left(\frac{T_g^2}{T^2} - 1\right)t\right]\} + 2b'}$$

where,

$$b' = \frac{1}{2} \left(1 - \frac{T_g^2}{T^2}\right)$$

Therefore, for very large as well as very small  $t$ , relaxation of  $Q$  is again exponential.



Both real experiments [76] and computer simulation [281] revealed that the relaxation is governed by either power law of the form

$$M(t) = M(0)t^{-a} \quad (4.18)$$

or by a logarithmic law

$$M(t) = M(0) - S(T)\ln t \quad (4.19)$$

However, it is difficult to distinguish power law (4.18) with  $a \ll 1$  from logarithmic law (4.19). We shall derive (4.19) using a DRT in Chapter 8.

The study of the self-correlation function  $R(t) = \langle S_i(t) S_i(0) \rangle_{av}$  involves setting up a Master equation for the conditional probability  $P(\dots, S_i, \dots; t \mid \dots, S_i', \dots, 0)$ . Proceeding exactly as before we get

$$\begin{aligned} \left(\frac{\eta}{k_B T}\right) \frac{dR(t)}{dt} = & - \left(1 - \frac{T_g^2}{T^2}\right) R(t) + \left(\frac{T_c}{T}\right) M(t) M(0) \\ & - \left(\frac{T_g^2}{T^2}\right) Q(t) R(t) \end{aligned} \quad (4.20)$$

The structure of equation (4.20) is exactly similar to (4.5) except for the factor  $(\eta/2k_B T)$  being replaced by  $(\eta/k_B T)$ . It is therefore easy to see that the equilibrium values for  $R$  and  $Q$  are the same. This is the justification for using  $Q(\infty)$  as the static spin glass order parameter. Hence, assuming  $M(0) = 0$ , the solutions for  $R(t)$  are

$$R(t) = R(0) \exp(-t/\tau_{g>}) \text{ for } T > T_g$$

and

$$R(t) = R(\infty) + [R(0) - R(\infty)] \exp(-t/\tau_{g<}) \text{ for } T < T_g.$$

Taking the Fourier transform and remembering that  $R(\infty) = Q$  we get,

$$R(\omega) = Q \delta(\omega) + [R(0) - Q] \frac{\tau_g / \pi}{\omega^2 \tau_g^2 + 1} \quad (4.21)$$

where,

$$\begin{aligned} \tau_g &= \tau_{g>} \quad \text{for } T > T_g \\ &= \tau_{g<} \quad \text{for } T < T_g. \end{aligned}$$

Notice that we have not used eigenstate space technique used by earlier authors. Besides, our treatment yields coupled relaxation of  $M$  and  $Q$ .

## 4.2 Resistivity :

In this section we shall apply equations (4.21) to calculate electrical resistivity in canonical spin glasses. We shall consider two separate sources which contribute to conduction electron scattering and hence to resistivity: s-electrons scattering against the potential fluctuations in the alloy and by localised impurity moments on the magnetic atoms. The hamiltonian of such a system may be written as

$$\begin{aligned}
H = H_0 + (1/N) \sum_{\underline{k}} \sum_{\underline{k}'} \sum_{\sigma} \sum_{\underline{r}_i} n_i V_{\underline{k}\underline{k}'} e^{i(\underline{k}-\underline{k}')\cdot\underline{r}_i} a_{\underline{k}'\sigma}^+ a_{\underline{k}\sigma} \\
- (1/N) \sum_{\underline{k}} \sum_{\underline{k}'} \sum_{\underline{r}_i} n_i J_{\underline{k}\underline{k}'} [(a_{\underline{k}'\uparrow}^+ a_{\underline{k}\uparrow} - a_{\underline{k}'\downarrow}^+ a_{\underline{k}\downarrow}) S_i^z \\
+ a_{\underline{k}'\downarrow}^+ a_{\underline{k}\uparrow} S_i^+ + a_{\underline{k}'\uparrow}^+ a_{\underline{k}\downarrow} S_i^-] e^{-i(\underline{k}-\underline{k}')\cdot\underline{r}_i} \quad (4.22)
\end{aligned}$$

$H_0$  is the Hamiltonian of the s-electrons in the host,  $V_{\underline{k}\underline{k}'}$  the potential difference between the host and the impurity,  $J_{\underline{k}\underline{k}'}$  is the s-d coupling between the conduction s-electrons and the moment carrying d-electrons of the magnetic impurities and  $n_i$  is the occupation probability of the site  $\underline{r}_i$  by a magnetic impurity. We shall consider both  $V_{\underline{k}\underline{k}'}$  and  $J_{\underline{k}\underline{k}'}$  to be isotropic  $V$  and  $J$ .

The transition probability  $\Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma')$  is given in the Born approximation [339]

$$\begin{aligned}
\Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma') &= f(\underline{k}\sigma) [1 - f(\underline{k}'\sigma')] \sum_{i,j} n_i n_j e^{i(\underline{k}-\underline{k}')\cdot(\underline{r}_i-\underline{r}_j)} \\
&\left( \frac{1}{2\pi N^2} \right) \int_{-\infty}^{\infty} d\omega \delta(\omega - \epsilon_{\underline{k}\sigma} + \epsilon_{\underline{k}'\sigma'}) \int_{-\infty}^{\infty} dt e^{i\omega t} \\
&[J^2 \langle S_i(t) \cdot S_j(0) \rangle + V^2 + \hat{\sigma} \cdot \hat{\sigma} JV \langle S_i^z \rangle] \quad (4.23)
\end{aligned}$$

where  $f$  is the Fermi function and  $\hat{\sigma}$  is the unit vector along  $\sigma$ . We shall calculate the spin flip and non spin flip scatterings separately.

(a) Non-spin Flip Scattering

In this case  $\sigma = \sigma'$ . Therefore, neglecting any short ranged order, the probability of transition is given by:

$$\sum_{\underline{k}'\sigma} \Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma) = A + B + C \quad (4.24)$$

where,

$$\begin{aligned} A &= \sum_{\underline{k}'\sigma} f(\epsilon_{\underline{k}}) \{1 - f(\epsilon_{\underline{k}'})\} \frac{J^2}{2\pi N^2} \\ &\sum_i n_i \int_{-\infty}^{\infty} d\omega \delta(\omega - \epsilon_{\underline{k}\sigma} + \epsilon_{\underline{k}'\sigma}) \int_{-\infty}^{\infty} dt \langle S_i^z(t) S_i^z(0) \rangle e^{i\omega t} \\ &= \frac{J^2}{N} \sum_i n_i \int_{-\infty}^{\infty} d\omega R_{zz}^i(\omega) \frac{1}{N} \sum_{\underline{k}'\sigma} f(\epsilon_{\underline{k}}) \{1 - f(\epsilon_{\underline{k}'})\} \\ &\quad \delta(\omega - \epsilon_{\underline{k}\sigma} + \epsilon_{\underline{k}'\sigma}) \end{aligned}$$

where,

$$R_{zz}^i(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle S_i^z(t) S_i^z(0) \rangle$$

$$\text{Using the relation } f(\epsilon_{\underline{k}}) \{1 - f(\epsilon_{\underline{k}} - \omega)\} = \frac{f(\epsilon_{\underline{k}}) - f(\epsilon_{\underline{k}} - \omega)}{1 - e^{\beta\omega}}$$

We get,

$$\begin{aligned} A &= \frac{J^2}{N} \sum_i n_i \int_{-\infty}^{\infty} d\omega R_{zz}^i(\omega) \left\{ \frac{f(\epsilon_{\underline{k}}) - f(\epsilon_{\underline{k}} - \omega)}{1 - e^{\beta\omega}} \right\} \\ &\quad \sum_{\sigma} N(\epsilon_{\underline{k}\sigma} - \omega) \end{aligned} \quad (4.25)$$

$$\begin{aligned}
B &= \sum_{\underline{k}', \sigma} f(\epsilon_{\underline{k}}) \{1 - f(\epsilon_{\underline{k}'})\} \frac{V^2}{N^2} \sum_i n_i \int_{-\infty}^{\infty} d\omega \\
&\quad \delta(\omega - \epsilon_{\underline{k}\sigma} + \epsilon_{\underline{k}'\sigma}) \int_{-\infty}^{\infty} dt e^{i\omega t} \\
&= \frac{V^2}{N} \sum_{\underline{k}', \sigma} f(\epsilon_{\underline{k}}) \{1 - f(\epsilon_{\underline{k}'})\} \frac{1}{N} \sum_i n_i \delta(\epsilon_{\underline{k}'\sigma} - \epsilon_{\underline{k}\sigma})
\end{aligned}$$

Using the relation  $f(\epsilon_{\underline{k}}) \{1 - f(\epsilon_{\underline{k}})\} = -\beta^{-1} \left( \frac{df}{d\epsilon_{\underline{k}}} \right)$

$$\begin{aligned}
B &= -\beta^{-1} \left( \frac{df}{d\epsilon_{\underline{k}}} \right) \frac{V^2}{N} \sum_i n_i \frac{1}{N} \sum_{\underline{k}', \sigma} \delta(\epsilon_{\underline{k}'\sigma} - \epsilon_{\underline{k}\sigma}) \\
&= \frac{V^2}{N} \left[ -\beta^{-1} \left( \frac{df}{d\epsilon_{\underline{k}}} \right) \sum_i n_i \sum_{\sigma} N(\epsilon_{\underline{k}\sigma}) \right] \quad (4.26)
\end{aligned}$$

Similarly,

$$C = \left( \frac{2JV}{N} \right) \left[ -\beta^{-1} \left( \frac{df}{d\epsilon_{\underline{k}}} \right) \sum_i n_i \langle S_i^z \rangle \sum_{\sigma} \partial N(\epsilon_{\underline{k}\sigma}) \right] \quad (4.27)$$

In equations (4.25) - (4.27)  $N(\epsilon_{\underline{k}\sigma})$  is the density of states of electrons with energy  $\epsilon_{\underline{k}}$  and spin  $\sigma$ . Since  $M = [\langle S_i^z \rangle]_{av} = 0$  in the spin glass phase  $C = 0$ . Now carrying out the configuration averaging, the total non-spin flip scattering contribution to the transition probability becomes

$$\begin{aligned}
\beta \left[ \sum_{\underline{k}', \sigma} \Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma) \right]_{av} &= - \left( \frac{df}{d\epsilon_{\underline{k}}} \right) c [V^2 N_+(\epsilon_{\underline{k}}) \\
&\quad - \frac{\beta J^2}{\left( \frac{df}{d\epsilon_{\underline{k}}} \right)} \int_{-\infty}^{\infty} d\omega]
\end{aligned}$$

$$R_{ZZ}(\omega) = N_+(\epsilon_{\underline{k}} - \omega) \left\{ \frac{f(\epsilon_{\underline{k}} - \omega) - f(\epsilon_{\underline{k}})}{e^{\beta\omega} - 1} \right\} \quad (4.28)$$

where  $c = \frac{1}{N} \sum_i n_i$  is the concentration of magnetic impurities,

$$N_+ = N(\epsilon_{\underline{k}\uparrow}) + N(\epsilon_{\underline{k}\downarrow})$$

(b) Spin Flip Scattering:

In this case  $\sigma \neq \sigma'$ , and

$$\Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma') = f(\epsilon_{\underline{k}\sigma}) \{1 - f(\epsilon_{\underline{k}'\sigma'})\} \frac{J^2}{N^2} \sum_i n_i$$

$$\int_{-\infty}^{\infty} d\omega \delta(\omega - \epsilon_{\underline{k}\sigma} + \epsilon_{\underline{k}'\sigma'}) \{R_{xx}^i(\omega) + R_{yy}^i(\omega)\}$$

Therefore,

$$\beta \left[ \sum_{\underline{k}'\sigma'} \Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma') \right]_{av} = - \left( \frac{df}{d\epsilon_{\underline{k}}} \right) c \left[ \frac{J^2 \beta}{-(df/d\epsilon_{\underline{k}})} \right]$$

$$\int_{-\infty}^{\infty} d\omega \{R_{xx}(\omega) + R_{yy}(\omega)\} N_+(\epsilon_{\underline{k}} - \omega) \left\{ \frac{f(\epsilon_{\underline{k}} - \omega) - f(\epsilon_{\underline{k}})}{e^{\beta\omega} - 1} \right\} \quad (4.29)$$

In the spin glass phase  $R_{xx}(\omega) = R_{yy}(\omega) = R_{zz}(\omega)$  because of average isotropy of the spin orientations. Now, using the relation that

$$-(df/d\epsilon_{\underline{k}}) \tau_{\underline{k}\sigma}^{-1} = \beta \left[ \sum_{\underline{k}'\sigma'} \Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma') \right]_{av}$$

and resistivity  $\rho = \left( \frac{m^*}{ne^2} \right) \tau_e^{-1}$  we have

calculated, using single relaxation time, shows cusp at  $T_g$ , contrary to the smooth behaviour observed experimentally. This is shown to be a consequence of the inadequacy of cooperative phase transition model with single relaxation time.

Chapter 5 is an extension of Chapter 4 to vector spin glasses in the presence of external magnetic field. In this Chapter we improve Mookerjee's theory of magnetoresistance incorporating effects of spin dynamics.

Part C of this thesis begins with Chapter 6 where we develop the percolation model of paramagnet-to-spin glass transition and calculate the transition temperature  $T_g$  as a function of concentration  $c$  taking the effects of finite mean free path and frustration properly into account. In this Chapter we derive Vogel-Fulcher law and explain the variation of  $T_g$  with time and length scales of probes. Besides, we calculate the low-field low-frequency a.c. susceptibility and show that linear response theory holds.

In Chapter 7 the mathematical relation between the percolation model and Wohlfarth's model is established and interpreted physically. Comparing our theoretical results with experimental data we calculate the distribution of blocking temperatures.

The logarithmic relaxation of magnetization is derived analytically in Chapter 8 so as to reveal a deeper connection with  $1/f$  noise in various other physical systems.

Part B highlights some further inadequacies of the cooperative phase transition model of Edwards-Anderson type with single relaxation time. Part C proposes an alternative theoretical framework whose predictions are, at least qualitatively, closer to the experimental results. The latter paves the way for an unified approach to the theories of spin glasses by incorporating cooperative effects on the one hand, and features of a gradual freezing process on the other.

Part B begins with Chapter 2 where we investigate the physical conditions under which both Lorentzian and Gaussian local field distributions are good approximations for Ising spin glasses with RKKY exchange interaction. In this chapter we also solve Sherrington-Kirkpatrick equations for two canonical spin glasses numerically and self-consistently so as to find the variation of the order parameters with temperature and external magnetic field.

Mookerjee's theory of magnetoresistance has been applied to two canonical spin glasses in Chapter 3.

Comparing the magnetoresistance thus calculated with the corresponding experimental data we calculate the strength of the s-d exchange interaction,  $J_{sd}$ .

The results of our investigation of relaxation spin dynamics in spin glasses using Master equation approach has been applied in Chapter 4 to calculate the resistivity by Boltzmann transport equation method. The resistivity thus



## SYNOPSIS

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Because of the exchange interaction among the moment bearing magnetic atoms (or ions) in random magnetic alloys, various kinds of magnetic ordering take place depending on the temperature and the concentration,  $c$ , of the magnetic constituent(s). In this thesis we have investigated the nature of the transition from paramagnet-to-spin glass state and the behaviour of various physical properties across the transition.

Recently there have been several attempts to regard a spin glass as the prototype for many more complicated and less understood physical systems. Unfortunately, even the theoretical understanding of the physics of spin glasses is far from complete. There have been two distinct and apparently irreconcilable approaches - the cooperative phase transition model and the model of gradual blocking of superparamagnetic clusters. Both of these have had limited success. All the successes and limitations of these models have been critically reviewed in part A of this thesis.

$$\tau_e^{-1} = c [V^2 N_+(\epsilon_F) - \frac{3\beta J^2}{(\frac{df}{d\epsilon})_{\epsilon_k=\epsilon_F}} \int_{-\infty}^{\infty} d\omega R(\omega) N_+(\epsilon_F - \omega) \left\{ \frac{f(\epsilon_k - \omega) - f(\epsilon_k)}{e^{\beta\omega} - 1} \right\}_{\epsilon_k=\epsilon_F}] \quad (4.30)$$

We have considered  $\epsilon_k \approx \epsilon_F$  because due to the derivative of the Fermi function in expression (4.30), it is clear that only the electrons near the Fermi surface contribute dominantly to resistivity. Assuming, further  $\omega$  to be small

$$f(\epsilon_F - \omega) - f(\epsilon_F) \approx \beta \omega / 4$$

which is true for reasonable values of  $\omega$  (note that  $\hbar = 1$  throughout),

$$\tau_e^{-1} = c [V^2 N_+(\epsilon_F) + 3\beta J^2 N_+(\epsilon_F) \int_{-\infty}^{\infty} \frac{\omega R(\omega)}{(e^{\beta\omega} - 1)} d\omega] \quad (4.31)$$

An equation exactly similar to (4.31) was derived by Campbell [348] from intuitive-qualitative arguments. Since,

$$R(\infty) = Q(T), \text{ and } R(0) = \frac{S(S+1)}{3} \quad (4.21)$$

can be written as

$$R(\omega) = Q(T) \delta(\omega) + \left[ \frac{S(S+1)}{3} - Q(T) \right] \frac{\gamma/\pi}{\omega^2 + \gamma^2}$$

where

$$\gamma = \tau_g^{-1} \quad \text{or} \quad \tau_g^{-1} \quad \text{according as } T > T_g$$

or  $T < T_g$ . Now onwards we shall use the symbol  $\tau$  for  $\tau_g$ . Then

$$\int_{-\infty}^{\infty} \frac{\omega R(\omega)}{e^{\beta\omega} - 1} d\omega = \frac{Q(T)}{\beta} + \left[ \frac{S(S+1)}{3} - Q(T) \right] \frac{\gamma}{\pi}$$

$$\int_{-\infty}^{\infty} \frac{\omega d\omega}{(e^{\beta\omega} - 1)(\omega^2 + \gamma^2)}$$

Let us first check the asymptotic behaviour of equation (4.31). At  $T = 0$ ,  $Q = S^2/3$  and  $\tau = 0$  i.e.  $\gamma = \infty$ . Besides as  $T \rightarrow 0$ ,  $\beta \rightarrow \infty$ . Hence in this limit

$$\frac{\beta\gamma}{\pi} \int_{-\infty}^{\infty} \frac{\omega d\omega}{(e^{\beta\omega} - 1)(\omega^2 + \gamma^2)} \cong \frac{\gamma}{\pi} \int_{-\infty}^{\infty} \frac{d\omega}{(1 + \frac{\beta\omega}{2!} + \frac{\beta^2\omega^2}{3!} + \dots)} = 0$$

and, we have

$$\rho(0) = \left( \frac{m^*}{ne^2} \right) c N_+(\epsilon_F) [V^2 + J^2 S^2] \quad (4.22)$$

which is the correct Yoshida limit [349]. On the other hand, as  $T \rightarrow \infty$ ,  $Q = 0$  and  $\tau \rightarrow 0$ . In this case,  $\beta\omega$  is small and we have

$$\frac{\beta\gamma}{\pi} \int_{-\infty}^{\infty} \frac{\omega d\omega}{(e^{\beta\omega} - 1)(\omega^2 + \gamma^2)} \cong \frac{\gamma}{\pi} \int_{-\infty}^{\infty} \frac{d\omega}{(\omega^2 + \gamma^2)} = 1$$

Hence,

$$\rho(\infty) = \left( \frac{m^*}{ne^2} \right) c N_+(\epsilon_F) [V^2 + J^2 S(S+1)] \quad (4.23)$$

which is, again, the correct Yoshida limit [349].

It is interesting to notice that the integral above is exactly the same as encountered in computing dynamical susceptibility in the microscopic theory of spin fluctuations

(see integral (3.8) of Ramakrishnan [350]). The integral may be split into two parts as follows:

$$\frac{\gamma}{\pi} \int_{-\infty}^{\infty} \frac{\beta \omega d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)} = \frac{\gamma}{\pi} \left[ \int_{-\infty}^0 \frac{\beta \omega d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)} + \int_0^{\infty} \frac{\beta \omega d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)} \right]$$

Now,

$$\int_{-\infty}^0 \frac{\beta \omega d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)} = \int_0^{\infty} \frac{\beta \omega d\omega}{(1 - e^{-\beta \omega})(\omega^2 + \gamma^2)}$$

$$= \int_0^{\infty} \frac{\beta \omega e^{\beta \omega} d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)} = \int_0^{\infty} \frac{(\beta \omega e^{\beta \omega} - \beta \gamma^2) d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)}$$

$$+ \int_0^{\infty} \frac{\beta \omega d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)} = \int_0^{\infty} \frac{\beta \omega d\omega}{(\omega^2 + \gamma^2)}$$

$$+ \int_0^{\infty} \frac{\beta \omega d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)}$$

i.e.,

$$\frac{\gamma}{\pi} \int_{-\infty}^{\infty} \frac{\beta \omega d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)} = \frac{\gamma}{\pi} \int_0^{\infty} \frac{\beta \omega d\omega}{(\omega^2 + \gamma^2)} + \frac{2\gamma}{\pi} \int_0^{\infty} \frac{\beta \omega d\omega}{(e^{\beta \omega} - 1)(\omega^2 + \gamma^2)}$$

The upper limit of integration in the first integral is taken to be  $\omega_M \approx 0$  ( $k_B T_g$ ). Following Ramakrishnan [350], we absorb the effect of this term in  $H_0$ . As a result  $H_0$  gets renormalized to  $H_0^{\text{eff}}$ . Hence, finally, resistivity is given by

$$\rho(T) = \left(\frac{m^*}{ne^2}\right) cN_+(\epsilon_F) \left[ V^2 + 3\beta J^2 \left[ \frac{Q(T)}{\beta} + \left\{ \frac{S(S+1)}{3} - Q \right\} \frac{\phi(T, \tau)}{\pi\tau} \right] \right] \quad (4.24)$$

where,

$$\phi(T, \tau) = \ln y - (2y)^{-1} - \Psi(y)$$

with  $y = \beta / 2\pi\tau$  and  $\Psi(y)$  is the digamma function.

Ramakrishnan [350] has given an approximate form for  $\phi(y)$  valid over a large range of  $y$ :

$$\phi(y) = (2y + 12y^2)^{-1} \quad (4.25)$$

Let us now investigate the behaviour of  $\rho(T)$  at some particular temperatures, viz.  $T = 0$ ,  $\infty$  and  $T = T_g$ . For brevity let us call

$$\frac{\beta \phi(T, \tau)}{\pi\tau} \cong \frac{\pi k_B T \tau}{\pi k_B T \tau + 3} = X(T)$$

(i) Very Low Temperature ( $T \rightarrow 0$ ):

As  $T \rightarrow 0$ ,  $T\tau \rightarrow 0$  and hence  $X(T) \rightarrow 0$ . Besides,  
 $Q(T = 0) = S^2/3$ .

Therefore,

$$\rho(0) = \left(\frac{m^*}{ne^2}\right) cN_+(\epsilon_F) [V^2 + J^2 S^2]$$

which agrees with the Yoshida limit (4.22). This agreement establishes the validity of the approximate form (4.25)

for  $T \ll T_g$ .

(ii) Very High Temperatures ( $T \rightarrow \infty$ ):

In this regime  $k_B T \tau = \frac{\eta}{(1 - \frac{T_g^2}{T^2})}$  and hence  $k_B T \tau \rightarrow \eta$

as  $T \rightarrow \infty$ .

Therefore  $X(T) \rightarrow \frac{\pi}{\pi + (3/\eta)}$  as  $T \rightarrow \infty$ .

Thus,

$$\rho(\infty) = \left(\frac{m^*}{ne^2}\right) cN_+(\epsilon_F) \left[ V^2 + \frac{\pi J^2 S(S+1)}{\pi + (3/\eta)} \right]$$

which does not agree with the Yoshida limit (4.23), and hence the inapplicability of the approximate form (4.25) for  $T \gg T_g$ .

(iii) As  $T \rightarrow T_g$  from both above and below,  $X(T) \rightarrow 1$  so that

$$\begin{aligned} \rho(T_g) &= \left(\frac{m^*}{ne^2}\right) cN_+(\epsilon_F) [V^2 + J^2 S(S+1)] \\ &= \rho(\infty). \end{aligned}$$

Let us now examine the slope of  $\rho(T)$  near  $T_g$ . We know that

$$\frac{d\tau}{dT} = \left( \frac{1}{T} - \frac{2k_B \tau_{>}}{\eta} \right) \tau_{>} \quad \text{for } T > T_g$$

and

$$\frac{d\tau}{dT} = \left( \frac{1}{T} + \frac{2k_B \tau_{<}}{\eta} \right) \tau_{<} \quad \text{for } T < T_g.$$

Since  $\frac{1}{T} \ll \frac{2k_B \tau}{\eta}$  for  $T$  near  $T_g$ ,

$$\frac{d\tau_{>}}{dT} = - \frac{2k_B \tau_{>}^2}{\eta} \quad \text{for } T > T_g$$

and

$$\frac{d\tau_{<}}{dT} = \frac{2k_B \tau_{<}^2}{\eta} \quad \text{for } T < T_g.$$

Thus the derivative of  $\tau$  is negative above  $T_g$  and positive below  $T_g$ . This is physically consistent because  $\tau_{>}$  increases from zero to infinity as the temperature is decreased from large values to  $T_g$ , while  $\tau_{<}$  decreases from infinity to zero as the temperatures decreases from  $T_g$  to zero. Notice that  $\tau_{<}$  is the lifetime of deviations from the frozen equilibrium configuration of spins below  $T_g$ .

Now,

$$\begin{aligned} & \frac{d}{dT} \left[ X \left\{ \frac{S(S+1)}{3} - Q \right\} + Q \right] \\ &= \left\{ \frac{S(S+1)}{3} - Q \right\} \frac{dX}{dT} + (1 - X) \frac{dQ}{dT} \\ &= \left[ \frac{\pi k_B \tau}{(\pi k_B T \tau + 3)} + \frac{\pi k_B T}{(\pi k_B T \tau + 3)} \cdot \left( \frac{d\tau}{dT} \right) \right. \\ & \quad \left. - \frac{\pi^2 k_B^2 T \tau^2}{(\pi k_B T \tau + 3)^2} - \frac{\pi^2 k_B^2 T^2 \tau}{(\pi k_B T \tau + 3)^2} \left( \frac{d\tau}{dT} \right) \right] \left\{ \frac{S(S+1)}{3} - Q \right\} \\ & \quad + (1 - X) \frac{dQ}{dT} \end{aligned}$$

$$= \left[ \frac{\pi k_B \tau}{(\pi k_B T \tau + 3)} + \frac{\pi k_B T \tau}{(\pi k_B T \tau + 3)} \left( \frac{1}{T} - \frac{2k_B \tau}{\eta} \right) - \frac{\pi^2 k_B^2 T^2 \tau^2}{(\pi k_B T \tau + 3)^2} - \frac{\pi^2 k_B^2 T^2 \tau^2}{(\pi k_B T \tau + 3)^2} \left( \frac{1}{T} - \frac{2k_B \tau}{\eta} \right) \right]$$

$$\left\{ \frac{S(S+1)}{3} - Q \right\} + (1 - X) \frac{dQ}{dT}.$$

$$= \left[ \frac{2\pi k_B T \tau}{(\pi k_B T \tau + 3)} \left( \frac{1}{T} - \frac{k_B \tau}{\eta} \right) - \frac{2\pi^2 k_B T^2 \tau^2}{(\pi k_B T \tau + 3)^2} \left( \frac{1}{T} - \frac{k_B \tau}{\eta} \right) \right]$$

$$\left\{ \frac{S(S+1)}{3} - Q \right\} + (1 - X) \frac{dQ}{dT}.$$

$$= \frac{2\pi k_B T \tau}{(\pi k_B T \tau + 3)} \left( \frac{1}{T} - \frac{k_B \tau}{\eta} \right) \left[ 1 - \frac{\pi k_B T \tau}{(\pi k_B T \tau + 3)} \right]$$

$$\left\{ \frac{S(S+1)}{3} - Q \right\} + (1 - X) \frac{dQ}{dT}.$$

$$= \frac{6\pi k_B T \tau}{(\pi k_B T \tau + 3)^2} \left( \frac{1}{T} - \frac{k_B \tau}{\eta} \right) \left[ \frac{S(S+1)}{3} - Q \right]$$

$$+ (1 - X) \frac{dQ}{dT}.$$

$$= - \frac{2S(S+1)}{\eta T_g \pi} \quad \text{for } T \rightarrow T_g \text{ from above as } X(T_g) = 1.$$

$$\text{Similarly, } \frac{d}{dT} \left[ X \left\{ \frac{S(S+1)}{3} - Q \right\} + Q \right] = \frac{2S(S+1)}{\eta T_g \pi}$$

for  $T \rightarrow T_g$  from below.



Therefore  $\frac{d\rho}{dT}$  exhibits a discontinuity at  $T = T_g$ . In other words (4.24) exhibits a cusp at  $T_g$ . Experimentally no such cusp has been observed so far in any of the canonical SG [80,341]. We attribute this discrepancy to the oversimplified picture of homogeneous MFT of cooperative phase transition model where there is a single relaxation time. We shall show in Chapter 9 how a distribution of relaxation times gives rise to better agreement with experimental results.

## CHAPTER 5\*

### SPIN DYNAMICS IN THE PRESENCE OF EXTERNAL MAGNETIC FIELD AND MAGNETORESISTANCE

In this Chapter we shall study the spin dynamics in the presence of external magnetic field along the same line as adopted in Chapter 4. The results of this investigation will be utilized to calculate the magnetoresistance of canonical SG.

#### 5.1 Spin Dynamics in the Presence of Field

Generalizing equations (4.4) and (4.5) to vector SG in the presence of external uniform magnetic field  $H$ , we get

$$\begin{aligned} \left(\frac{\eta}{k_B T}\right) \frac{dR_{\mu\nu}(t)}{dt} = & - \left(1 - \frac{T^2}{T^2}\right) R_{\mu\nu}(t) + \left(\frac{T}{T}\right) M_\mu(t) M_\nu(0) \\ & - \left(\frac{T^2}{T^2}\right) Q_{\mu\nu}(t) R_{\mu\nu}(t) - \left(\frac{\gamma\eta}{k_B T}\right) [\langle \underline{S}(t) \times \underline{H} \rangle_\mu S_\nu(C)]_{av} \end{aligned} \quad (5.1)$$

where  $R_{\mu\nu}(t) = [\langle S_\mu^u(t) S_\nu^v(0) \rangle]_{av}$

and,

$$\begin{aligned} \left(\frac{\eta}{k_B T}\right) \frac{dM_\mu(t)}{dt} = & - \left(1 - \frac{T}{T}\right) M_\mu(t) - \frac{1}{3} \left(\frac{T}{T}\right)^3 M_\mu^3(t) \\ & - \left(\frac{T}{T^3}\right) M_\mu(t) Q_{\mu\mu}(t) + \left(\frac{\gamma\eta}{k_B T}\right) (\underline{M}(t) \times \underline{H})_\mu \end{aligned} \quad (5.2)$$

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because the precessional motion of a spin  $\underline{S}$  in an external magnetic field  $\underline{H}$  is given by  $\frac{d\underline{S}}{dt} = \gamma(\underline{S} \times \underline{H})$ , where  $\gamma$  is the gyromagnetic ratio.

Next we solve equations (5.1) and (5.2) in the low concentration regime ( $T_c < T_g$ ) at high temperatures ( $T > T_g > T_c$ ) and low temperatures ( $T_g > T > T_c$  and  $T_g > T_c > T$ ). In the high temperature regime we can drop the third term on the right hand side of equation (5.1) and second and third terms on the right hand side of equation (5.2). Let us assume that the external field is applied along the z-direction. Then the solutions for magnetization are given by

$$\begin{aligned} \begin{Bmatrix} M_x(t) \\ M_y(t) \end{Bmatrix} &= \begin{Bmatrix} M_x(0) \\ M_y(0) \end{Bmatrix} \cos(\gamma H t) \exp(-t/\tau_c) \\ &+ \begin{Bmatrix} M_y(0) \\ -M_x(0) \end{Bmatrix} \sin(\gamma H t) \exp(-t/\tau_c) \end{aligned} \quad (5.3)$$

and,

$$M_z(t) = M_z(\infty) + [M_z(0) - M_z(\infty)] \exp(-t/\tau_c) \quad (5.4)$$

where  $M_z(\infty)$  is the equilibrium value of  $M_z$ . Notice that  $M_x(\infty)$  and  $M_y(\infty)$  both vanish. The physical interpretations of equations (5.3) and (5.4) are very clear - the precessional motion of the magnetization vector is described by the sine and cosine factors in these expressions whereas the thermal decay is expressed by the exponential factor involving only one characteristic temperature  $T_c$ . Similarly, solving (5.2)

we get,

$$\begin{aligned}
 \begin{Bmatrix} R_{xx}(t) \\ R_{yx}(t) \end{Bmatrix} &= \begin{Bmatrix} R_{xx}(0) \\ R_{yx}(0) \end{Bmatrix} \cos(\gamma H t) \exp(-t/\tau_g) \\
 &+ \begin{Bmatrix} R_{yx}(0) \\ -R_{xx}(0) \end{Bmatrix} \sin(\gamma H t) \exp(-t/\tau_g) \\
 &+ DM_x(0) \begin{Bmatrix} M_x(t) \\ M_y(t) \end{Bmatrix} \left[ 1 - \exp\left(-\frac{k_B T_c}{\eta D} t\right) \right] \quad (5.5)
 \end{aligned}$$

where,

$$D^{-1} = \left(1 - \frac{T_g^2}{T T_c}\right)$$

$$\begin{aligned}
 \begin{Bmatrix} R_{xy}(t) \\ R_{yy}(t) \end{Bmatrix} &= \begin{Bmatrix} R_{xy}(0) \\ R_{yy}(0) \end{Bmatrix} \cos(\gamma H t) \exp(-t/\tau_g) \\
 &+ \begin{Bmatrix} R_{yy}(0) \\ -R_{xy}(0) \end{Bmatrix} \sin(\gamma H t) \exp(-t/\tau_g) \\
 &+ DM_y(0) \begin{Bmatrix} M_x(t) \\ M_y(t) \end{Bmatrix} \left[ 1 - \exp\left(-\frac{k_B T_c}{\eta D} t\right) \right] \quad (5.6)
 \end{aligned}$$

and,

$$\begin{aligned}
 R_{zz}(t) &= R_{zz}(0) \exp(-t/\tau_g) \\
 &+ DM_z(0) M_z(t) \left[ 1 - \exp\left(-\frac{k_B T_c}{\eta D} t\right) \right] \quad (5.7)
 \end{aligned}$$

In the low temperature regime we write  $R_{\mu\nu}(t) = R_{\mu\nu}(\infty) + r_{\mu\nu}(t)$  and  $M_\mu(t) = M_\mu(\infty) + m_\mu(t)$  and get, as in Chapter 4,

$$\begin{aligned} \left(\frac{\eta}{k_B T}\right) \frac{dr_{\mu\nu}(t)}{dt} = & - \left(\frac{T_g^2}{T^2} - 1\right) r_{\mu\nu}(t) + \left(\frac{T_c}{T}\right) m_\mu(t) m_\nu(0) \\ & + \left(\frac{\gamma\eta}{k_B T}\right) \{ [\langle (S(t) \times H)_\mu S_\nu(0) \rangle]_{av} - [\langle (S(\infty) \times H)_\mu S_\nu(0) \rangle]_{av} \} \end{aligned} \quad (5.8)$$

and,

$$\begin{aligned} \left(\frac{\eta}{k_B T}\right) \frac{dm_\mu(t)}{dt} = & - \left(1 - \frac{2T_c}{T} + \frac{T_c T_g^2}{T^3}\right) m_\mu(t) \\ & + \left(\frac{\gamma\eta}{k_B T}\right) (m(t) \times H)_\mu \end{aligned} \quad (5.9)$$

Equations (5.8) and (5.9) can be solved exactly as equations (5.1) and (5.2). So the relaxation of  $R$  turns out to be exponential at all but one temperature. At  $T = T_g$ ,  $R_{\mu\nu}(t) \sim t^{-1/2}$  for all  $\mu, \nu$  at not too short time scales, exactly as in Chapter 4.

Unfortunately, the experimentally observed decay of  $\underline{M}$  and  $\underline{R}$  are much slower than exponential (probably logarithmic) and do not agree with the solutions obtained above. The correct behaviour will be derived in Chapter 8.

Let us now calculate  $R(\omega)$ . Proceeding exactly as in Chapter (4) and using the forms of  $\underline{R}(t)$  derived in this Chapter we get,

$$\begin{aligned}
R_{xx}(\omega) &= R_{yy}(\omega) = Q(T) \delta(\omega) \\
&+ \frac{\{ \frac{S(S+1)}{3} - Q(T) \}}{2 \pi \tau} \left[ \frac{1}{(\frac{1}{\tau})^2 + (\omega + \gamma H)^2} \right] \\
&- \frac{\{ \frac{S(S+1)}{3} - Q(T) \}}{2 \pi \tau} \left[ \frac{1}{(\frac{1}{\tau})^2 + (\omega - \gamma H)^2} \right]
\end{aligned} \quad (5.10)$$

$$R_{zz}(\omega) = Q(T) \delta(\omega) + \frac{\{ \frac{S(S+1)}{3} - Q(T) \}}{\pi \tau} \left[ \frac{1}{(\frac{1}{\tau})^2 + \omega^2} \right] \quad (5.11)$$

$$R_{xy}(\omega) = - R_{yx}(\omega) = \frac{i S(S+1)}{6 \pi \tau} \left[ \frac{1}{(\frac{1}{\tau})^2 + (\omega - \gamma H)^2} - \frac{1}{(\frac{1}{\tau})^2 + (\omega + \gamma H)^2} \right] \quad (5.12)$$

Notice that (5.10) - (5.12) reduce to the correct limits for  $H = 0$  derived in Chapter 4.

## 5.2 Magnetoresistance

We proceed exactly as in Chapter 4. For non spin flip scattering ( $\sigma = \sigma'$ )

$$\sum_{\underline{k}'} \overline{f}(\underline{k} \rightarrow \underline{k}' \sigma) = A + B + C \quad (5.13)$$

where,

$$A = (J^2/N) \sum_i n_i \int d\omega R_{zz}^i(\omega) \left\{ \frac{f(\epsilon_{\underline{k}}) - f(\epsilon_{\underline{k}} - \omega)}{1 - e^{\beta\omega}} \right\} N(\epsilon_{\underline{k}\sigma} + \omega) \quad (5.14)$$

$$B = (V^2/N) \{ -\beta^{-1} \left( \frac{df}{d\varepsilon_{\underline{k}}} \right) \sum_{\underline{i}} n_{\underline{i}} N(\varepsilon_{\underline{k}\sigma}) \} \quad (5.15)$$

$$C = (2J V/N) \{ -\beta^{-1} \left( \frac{df}{d\varepsilon_{\underline{k}}} \right) \sum_{\underline{i}} n_{\underline{i}} \langle S_{\underline{i}}^z \rangle_{\hat{\sigma}} N(\varepsilon_{\underline{k}\sigma}) \} \quad (5.16)$$

Carrying out the configuration averaging

$$\begin{aligned} \left[ \sum_{\underline{k}'} \Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma) \right]_{av} &= - \left( \frac{df}{d\varepsilon_{\underline{k}}} \right) \beta^{-1} c \left[ V^2 N(\varepsilon_{\underline{k}\sigma}) \right. \\ &+ 2 \hat{\sigma} J V M(\varepsilon_{\underline{k}\sigma}) - J^2 \beta \left( \frac{df}{d\varepsilon_{\underline{k}}} \right)^{-1} \int_{-\infty}^{\infty} R_{zz}(\omega) \\ &\left. N(\varepsilon_{\underline{k}\sigma} + \omega) \left\{ \frac{f(\varepsilon_{\underline{k}} - \omega) - f(\varepsilon_{\underline{k}})}{e^{\beta\omega} - 1} \right\} d\omega \right] \end{aligned} \quad (5.17)$$

Next we consider spin-flip-scattering. We define,

$$R_{+-}(t) = [\langle S_{\underline{i}}^+(t) S_{\underline{i}}^-(0) \rangle]_{av}$$

and,

$$R_{-+}(t) = [\langle S_{\underline{i}}^-(t) S_{\underline{i}}^+(0) \rangle]_{av}$$

where,

$$S_j^{\pm} = S_j^x \pm i S_j^y$$

Notice that  $R_{+-}$  is associated with down  $\rightarrow$  up spin flip scattering, whereas  $R_{-+}$  is with up  $\rightarrow$  down. Hence, taking the conservation of energy into account, we get

$$\Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma') = f(\underline{k}\sigma) \{1 - f(\underline{k}'\sigma')\} (J^2/2N^2) \sum_i n_i$$

$$\int_{-\infty}^{\infty} d\omega [R_{+-}^i(\omega) \delta(\omega - \varepsilon_{\underline{k}\sigma} + \varepsilon_{\underline{k}'\sigma'} - \gamma H) \\ + R_{-+}^i(\omega) \delta(\omega - \varepsilon_{\underline{k}\sigma} + \varepsilon_{\underline{k}'\sigma'} + \gamma H)]$$

Hence,

$$\left[ \sum_{\underline{k}'} \Gamma(\underline{k}\sigma \rightarrow \underline{k}'\sigma') \right]_{\text{av}} = -\frac{1}{2} \left( \frac{df}{d\varepsilon} \right)_{\underline{k}} [J^2 c / (-\frac{df}{d\varepsilon})] N(\varepsilon_{\underline{k}\sigma}) \\ \int_{-\infty}^{\infty} d\omega \left\{ \frac{f(\varepsilon_{\underline{k}} - \gamma H) - f(\varepsilon_{\underline{k}})}{e^{\beta(-\gamma H)} - 1} R_{+-}(\omega) + \frac{f(\varepsilon_{\underline{k}} - \omega - \gamma H) - f(\varepsilon_{\underline{k}})}{e^{\beta(\omega + \gamma H)} - 1} R_{-+}(\omega) \right\} \quad (5.18)$$

Now onwards we shall consider  $\varepsilon_{\underline{k}} \sim \varepsilon_F$  because only the Fermi electrons contribute most dominantly in resistivity,

$$R_{+-}(\omega) = R_{xx}(\omega) + R_{yy}(\omega) + i \{R_{yx}(\omega) - R_{xy}(\omega)\}$$

and,

$$R_{-+}(\omega) = R_{xx}(\omega) + R_{yy}(\omega) - i \{R_{yx}(\omega) - R_{xy}(\omega)\}$$

Therefore,

$$R_{+-}(\omega) = 2Q(T) \delta(\omega) + \frac{2S(S+1)}{3\pi\tau} \left\{ \frac{1}{(\frac{1}{\tau})^2 + (\omega - \gamma H)^2} \right\} \\ - \frac{Q}{\pi\tau} \left\{ \frac{1}{(\frac{1}{\tau})^2 + (\omega - \gamma H)^2} + \frac{1}{(\frac{1}{\tau})^2 + (\omega + \gamma H)^2} \right\} \quad (5.19)$$

and,



$$R_{-+}(\omega) = 2Q(T)\delta(\omega) + \frac{2S(S+1)}{3\pi\tau} \left\{ \frac{1}{\left(\frac{1}{\tau}\right)^2 + (\omega+\gamma H)^2} \right\} \\ - \frac{Q}{\pi\tau} \left\{ \frac{1}{\left(\frac{1}{\tau}\right)^2 + (\omega+\gamma H)^2} + \frac{1}{\left(\frac{1}{\tau}\right)^2 + (\omega-\gamma H)^2} \right\} \quad (5.20)$$

Now,

$$I_1 = \int_{-\infty}^{\infty} d\omega R_{-+}(\omega) \left\{ \frac{f(\epsilon_F - \omega + \gamma H) - f(\epsilon_F)}{e^{\beta(\omega - \gamma H)} - 1} \right\} \\ = 2Q(T) \left\{ \frac{f(\epsilon_F + \gamma H) - f(\epsilon_F)}{(e^{-\beta\gamma H} - 1)} \right\} \\ + \frac{2S(S+1)}{3\pi\tau} \int_{-\infty}^{\infty} d\omega \left[ \frac{f(\epsilon_F + \gamma H - \omega) - f(\epsilon_F)}{e^{\beta(\omega - \gamma H)} - 1} \right] \left\{ \frac{1}{\left(\frac{1}{\tau}\right)^2 + (\omega - \gamma H)^2} \right\} \\ - \frac{Q}{\pi\tau} \int_{-\infty}^{\infty} d\omega \left[ \frac{f(\epsilon_F + \gamma H - \omega) - f(\epsilon_F)}{e^{\beta(\omega - \gamma H)} - 1} \right] \left\{ \frac{1}{\left(\frac{1}{\tau}\right)^2 + (\omega - \gamma H)^2} \right. \\ \left. + \frac{1}{\left(\frac{1}{\tau}\right)^2 + (\omega + \gamma H)^2} \right\} \quad (5.21)$$

First three integrals in (5.21) can be carried out analytically, as was done in Chapter 4, assuming  $\omega \pm \gamma H$  to be small. The fourth integral can be written as

$$\int_{-\infty}^{\infty} d\omega \left[ \frac{f(\epsilon_F - \omega + \gamma H) - f(\epsilon_F)}{e^{\beta(\omega - \gamma H)} - 1} \right] \left\{ \frac{1}{\left(\frac{1}{\tau}\right)^2 + 4\gamma H\omega + (\omega - \gamma H)^2} \right\}$$

In order to do this integral analytically, we approximate it by

$$\int_{-\infty}^{\infty} d\omega \left[ \frac{f(\epsilon_F - \omega + \gamma H) - f(\epsilon_F)}{e^{\beta(\omega - \gamma H)} - 1} \right] \left\{ \frac{1}{\left(\frac{1}{\tau}\right)^2 + (\omega - \gamma H)^2} \right\}$$

where,

$$\tau' = \left[ \left(\frac{1}{\tau}\right)^2 + 4\gamma H \omega_0 \right]^{-\frac{1}{2}}$$

where we have replaced  $\omega$  within the bracket by a constant  $\omega_0$  independent of  $\omega$ . In this approximation,

$$\begin{aligned} \frac{1}{2} I_1 &= \frac{Q}{\beta} + \left\{ \frac{S(S+1)}{3\pi\tau} \right\} \phi(T, \tau) \\ &- \left( \frac{Q}{2\pi\tau} \right) \{ \phi(T, \tau) + \phi(T, \tau') \} \end{aligned} \quad (5.22)$$

where  $\phi(T, \tau)$  has the same approximate form (4.25). Similarly,

$$I_2 = \int_{-\infty}^{\infty} d\omega R_{-+}(\omega) \left\{ \frac{f(\epsilon_F - \omega - \gamma H) - f(\epsilon_F)}{e^{\beta(\omega + \gamma H)} - 1} \right\}$$

Therefore,

$$\frac{1}{2} I_2 = \frac{Q}{\beta} + \left\{ \frac{S(S+1)}{3\pi\tau} \right\} \phi - \frac{Q}{2\pi\tau} [\phi(T, \tau) + \phi(T, \tau'')] \quad (5.23)$$

where,

$$\tau'' = \left[ \left(\frac{1}{\tau}\right)^2 - 4\tau H \omega \right]^{\frac{1}{2}}$$

Notice that in the limit  $H = 0$ ,  $\tau' = \tau'' = \tau$ .

Using the approximate form (4.25) for  $\phi(T, \tau)$  we get

$$\varnothing(T, \tau') + \varnothing(T, \tau'') \approx \frac{3 k_B^3 T^3 (\tau'^2 \tau'' + \tau''^2 \tau') + 3 \pi^2 k_B^2 T^2 (\tau'^2 + \tau''^2)}{\pi^2 k_B^2 T^2 \tau' \tau'' + 3 \pi k_B T (\tau' + \tau'') + 9} \quad (5.24)$$

In order to compare theory with experiment we make a further approximation:

$$4 \gamma H \omega_0 \tau^2 \ll 1$$

so that retaining only terms upto the order of  $H$ ,

$$\tau' = (1 - 2 \gamma H \omega_0 \tau^2)$$

and,

$$\tau'' = (1 + 2 \gamma H \omega_0 \tau^2)$$

and hence,

$$\varnothing(T, \tau') + \varnothing(T, \tau'') = 2\varnothing(T, \tau)$$

Hence under this approximation, resistivity is given by [349]

$$\rho(H) \propto \left[ \frac{1}{W_0 + aM} + \frac{1}{W_0 - aM} \right]^{-1}$$

where,

$$W_0 = c V^2 N (\epsilon_F) + 3c\beta J^2 N (\epsilon_F) \left[ \frac{Q(H)}{\beta} + \frac{S(S+1)}{3 \pi \tau} \varnothing(T, \tau) - \frac{Q(H)}{\pi \tau} \varnothing(T, \tau) \right]$$

and,

$$a = 2 c J V N (\epsilon_F)$$

Therefore,

$$\rho(H) \propto \left[ W_0 - \left( \frac{a}{W_0} \right) M^2 \right] \quad (5.25)$$

As  $T \rightarrow 0$ ,  $\tau \rightarrow 0$  and hence  $Y \rightarrow 1$ , and as  $T \rightarrow T_g$ ,  $\tau \rightarrow \infty$ , hence  $Y \rightarrow 0$ . For very large  $T$ ,  $\tau \rightarrow 0$ , and again  $Y \rightarrow 1$ . Besides  $\Delta Q \rightarrow 0$  for both  $T \rightarrow 0$  and  $T \rightarrow \infty$ . Thus  $(4 M^2 - 3 \Delta Q Y) > 0$  for all  $T$ , and since terms of the order  $\frac{J^4}{V^2}$  are very small.  $\Delta \rho$  is negative at all  $T$ , in agreement with experiment and its value determined dominantly by  $(4 M^2 - 3 \Delta Q)$ .

Using the self-consistent solutions for  $M(H)$  and  $Q(H)$  obtained in Chapter 2 we plot  $(4 M^2 - 3 \Delta Q)$  against  $M^2$  for fixed temperatures in Figure 21. Thus, clearly  $\Delta \rho$  is proportional to  $M^2$  at least upto 12 KG. This in excellent agreement with experimental results [385, 386]. Besides, plotting  $(4 M^2 - 3 \Delta Q)$  against  $T$  for fixed  $H$  we obtained qualitative agreement with Nigam and Majumdar's [83] experimental data which show a maximum near  $T_g$ .

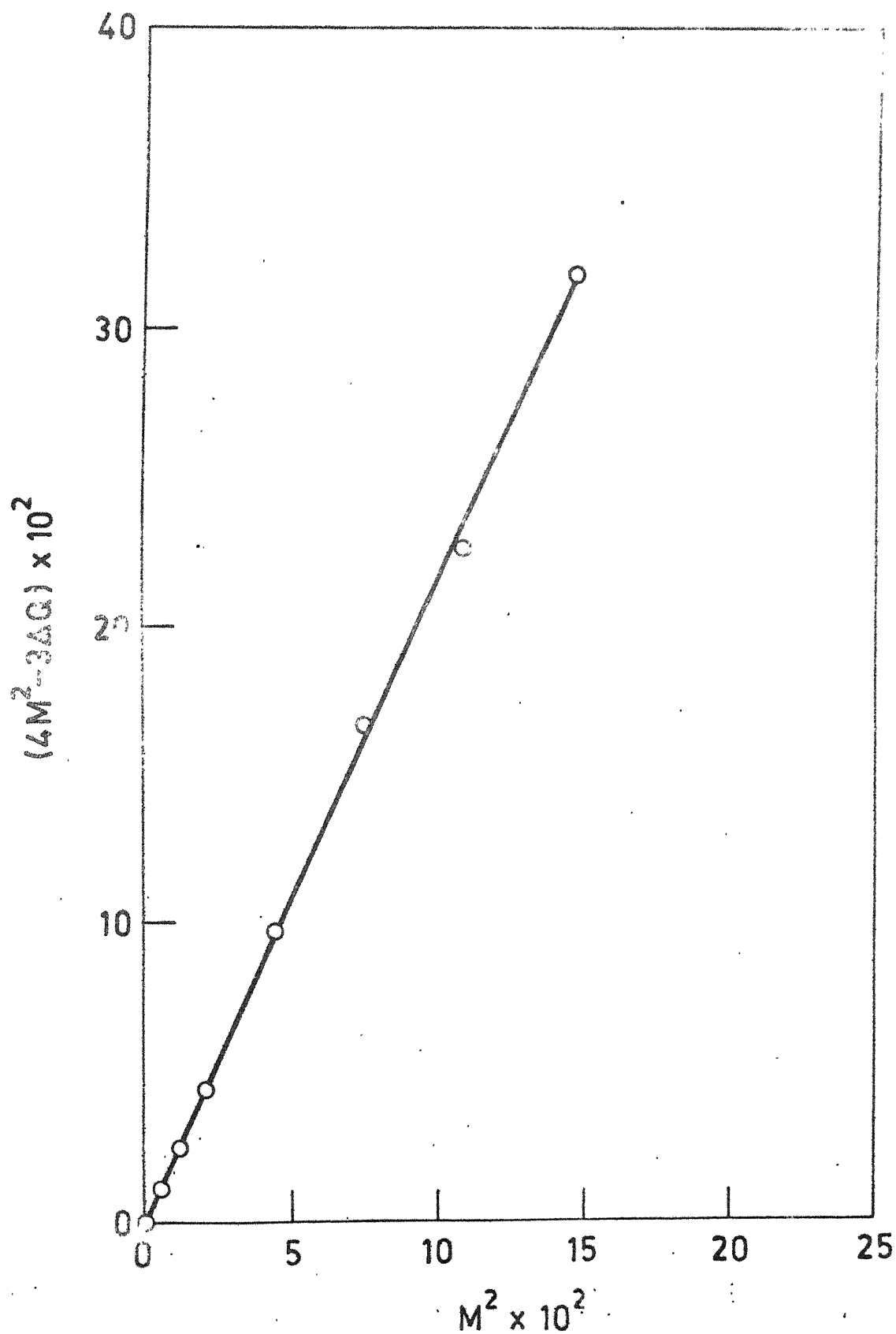


Figure 21 :  $(4M^2 - 3\Delta Q)$  for CuMn (0.7 at.%) plotted against  $M^2$  at a fixed temperature, 4.2K.

PART C

THE PERCOLATION MODEL  
OF  
SPIN GLASS TRANSITION

## CHAPTER 6<sup>\*</sup>

### SPIN GLASS TRANSITION AS A PERCOLATION PROBLEM

In this chapter we develop the basic framework of our percolation model [351]. Let us first describe the 'mechanism' of formation of 'locked' spin clusters. A free spin in contact with a heat bath, maintained at a temperature  $T$ , has a finite relaxation time  $t_0$ . For CuMn an estimate of  $t_0 = 10^{-13}$  sec. has been obtained from remanence data [352]. The presence of a second spin interacting with the first has a retardation effect changing the relaxation time. So long as  $J < k_B T$ , these two spins can be regarded as 'independent', albeit with lifetimes longer than a free spin. However, if  $J > k_B T$ , then in whichever configuration we begin, the spins eventually 'align' (either ferromagnetically or antiferromagnetically depending on the sign of  $J$ ) and from then onwards move as a single entity locked together. We may, therefore, define the locking criterion as  $J > k_B T$ , and hence the locking temperature

$$T_L = J/k_B$$

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\* Contents of this chapter have been published in J. Phys. F13, 431 (1983).

For RKKY interaction, the locking condition becomes

$$AR^{-3} \exp(-R/\delta) \geq k_F T \quad (6.1)$$

where  $R$  is the distance between the spins, and

$$A = \frac{J_{sd}^2}{\epsilon_F} \frac{(k_F a_0/2)^6}{\pi^3} \frac{\cos(2k_F R)}{(2k_F)^3}$$

and

$$\delta = \frac{\hbar k_F}{m^*},$$

$$a_0^3 = \text{volume of unit cell} = V_c$$

$$\epsilon_F = \frac{\hbar^2 k_F^2}{2m^*},$$

$$k_F = \text{Fermi momentum, and}$$

$$m^* = \text{effective mass of electrons.}$$

The exponential damping factor arises due to the scattering of the conduction electrons in a disordered system. If the dominant scatterers are the spins themselves, rather than other defects the damping is called self damping, and

$$\delta \propto c^{-1}.$$

On the other hand, for damping introduced by defects,  $\delta$  is independent of concentration [148]. We assume that our system remains metallic and  $\delta^3 \gg V_c$ .

This criterion (6.1) itself is insufficient to define the locking of two spins [40]. We have not yet taken



frustration into account. In this chapter, for the first time, effect of frustration will be incorporated into the percolation model of spin glass transition.

Because of the oscillating nature of the RKKY interaction and random occupation of the lattice sites by the magnetic constituents, the geometry of the locking bonds inside a locked cluster may lead to frustrated plaquettes. The frustrated bonds weaken 'locking' because the terminal spins, due to frustration, can still flip independently even if condition (6.1) is satisfied [40]. Moreover, a set of 'loose' spins may also exist even inside the locking sphere, if the local field that 'pins' the spin vanishes at some sites. We shall assume that such spins constitute a negligibly small fraction. This assumption will be justified in Chapter 11. Thus, in addition to condition (6.1) our criterion for the formation of a locking bond involves the examination of the bond topology locked according to condition (6.1), and the exclusion of all frustrated bonds from the cluster.

Next we geometrize the problem exactly as done by Smith [353]. Attached to every magnetic site is a sphere of radius  $\frac{1}{2}R$  where  $R$  is determined by the equality in condition (6.1). Two sites are bonded by a locking bond if their spheres overlap, and the environment of the bond is such that it does not form part of a frustrated plaquette. An  $n$ -cluster consists of a set of  $n$  such sites, each site is bonded to at least one other site within the set and no site is locked to any other site outside the set. In such

clusters, unlike ferromagnetic or antiferromagnetic clusters, all the spins do not necessarily align parallel or antiparallel, but depend upon the sign of the local magnetic field. If we try to flip a spin within a cluster, all the other spins in it try to flip with it. Since the sites may be distributed at random, amorphous as well as crystalline substitutional alloys may be taken into account.

As the temperature is reduced, the 'clusters' grow in size; more and more smaller 'clusters' coalesce together to form bigger clusters. At a critical radius, (i.e. at a critical temperature  $T_g$ ) an infinite cluster of locked spins first appears and percolation is said to have taken place.  $T_g$  is identified with the paramagnet to spin glass transition temperature. Such percolation process, where the concentration of sphere centres is kept fixed while the radius is varied, has been studied numerically by Kertesz [354]. Other kinds of percolation of spheres have been reviewed in our paper [351].

### 6.1 $T_g$ versus $c$ :

The crucial quantity of interest in the study of this percolation is the volume fraction

$$p(T) = 1 - \exp \left[ - \frac{4\pi}{3} \left\{ \frac{R^3(T)}{8} \right\} n \right] \quad (6.2)$$

where  $n$  is the number density of participating (non frustrated) sphere centres. Let  $n_0$  be the number density of all

atomic sites and  $c'$  be the fraction of these atoms that are magnetic and non-frustrated. Then  $n = n_0 c'$ . If  $c$  is small, frustration is small so that  $c' = c$ . As  $c$  increases but remains below  $c_p$  (percolation threshold for long-range magnetic order) frustration increases and  $c'$  becomes progressively less than  $c$ . We shall examine  $c' = f(c)$  as a function of  $c$  subsequently. From equations (6.1) and (6.2)

$$AR^{-3}(T_g) \exp[-R(T_g)/\delta] = k_B T_g \quad (6.3a)$$

and

$$y = R^3 c' \quad (6.3b)$$

where

$$y = \frac{6}{\pi n_0} \ln[1 - p(T_g)]$$

Eliminating  $R$  from (6.3a) and (6.3b) we get  $T_g$  as a function of  $c$ . We shall examine the relation between  $T_g$  and  $c$  in the following special cases.

Case (i): Infinite Mean Free Path and Very Low Concentration:

In this case

$$AR^{-3}(T_g) = k_B T_g$$

and

$$y = R^3 c,$$

because  $\delta \rightarrow \infty$  and  $c' = c$ . Hence  $y = Ac/k_B T_g$  implying  $T_g \propto c$ . This is in agreement with the numerical data of

Kinzel [354] and the scaling ideas of Souletie and Tournier [355]. In fact, the scaling laws of Souletie and Tournier follow from two facts —  $\frac{1}{r^3}$  law of interaction and  $c \rightarrow 0$  [108]. Both these facts have been utilized in case (i) and hence the agreement is not so surprising.

Case (ii) : Infinite Mean Free Path and  $c' < c$ :

In this case, exactly as in the earlier one

$$T_g \propto c'.$$

Thus frustration lowers  $T_g$  for a given  $c$ , because, less magnetic sites are available for cluster formation; consequently  $R(T_g)$  has to be larger or  $T_g$  lower than the value when frustration is absent and all magnetic sites participate in cluster formation.

Case (iii) : No Frustration ( $c' = c$ ) and Finite Mean Free Path:

Now,

$$\frac{Ac}{y} \exp[-y^{1/3} c^{-1/3}/\delta] = k_B T_g$$

Hence for self damping, (i.e.  $\delta \propto c^{-1}$ )

$$T_g \propto c \exp[-b c^{2/3}]$$

Thus, both frustration and finite mean free path lead to a weaker than linear concentration dependence of  $T_g$ . However, there is no evidence of a unique power law of the form  $T_g \propto c^b$  which holds across the whole concentration range.

Moreover, the functional dependence is not universal, as  $c'$  and  $\delta$  both depend on the structural and chemical nature of the system under consideration. This is contrary to the universal  $T_g \propto c^{1/2}$  law obtained in the mean field theories of Edwards and Anderson [114]. Real experiments on canonical spin glasses yield  $\beta$  varying between  $2/3$  to  $3/4$  [73]. Thus, our prediction turns out to be closer to experimental results.

From equations (6.3) we may write

$$T_g(c, \delta) = T_g(c, \infty) \exp[-R(c, T_g)/\delta]$$

where  $R(c, T_g)$  is the critical radius for a given concentration. This immediately gives us a direct physical interpretation of the 'characteristic length'

$$\phi(c) = - \frac{d(\ln T_g)}{d(1/\delta)} \quad (6.4)$$

of Kinzel [354] and Kinzel and Fischer [356].

Using  $p(T_g) = 0.28$  as obtained by Kertesz [354] and equations (6.1), (6.2) and (6.4) we get

$$\left(\frac{n_o r_n^3}{8}\right) c' = 0.07842 \times \frac{1}{[\phi(c)/r_n]^3} \quad (6.5)$$

For a given  $c$ , we put  $\phi(c)/r_n$  for  $\underline{\text{CuMn}}$  from Kinzel's data into equation (6.5) and evaluate  $c'$ . Figure 22 shows  $c'$  plotted against  $c$ . The behaviour confirms our earlier analysis.

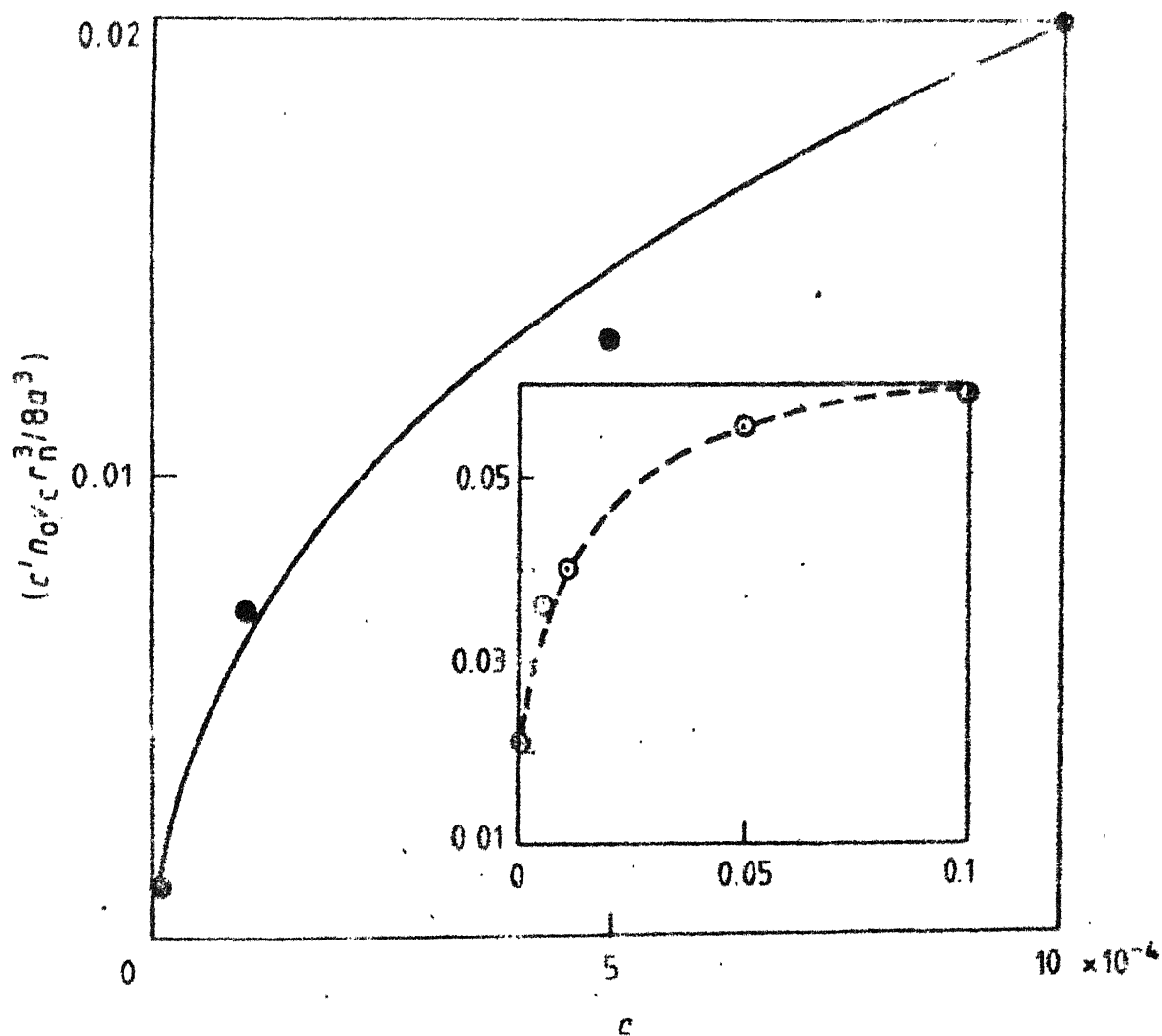


Figure 22 : Plot of  $c' = 1/c$  against  $c$ , obtained from the interpretation of the numerical data of Kinzel [354]. The units of  $c'$  are  $(n_0 v_c r_n^3 / 8a^3)$ ,  $n_0$  and  $v_c$  are explained in the text,  $r_n$  and  $a$  are as used by Kinzel.

## 6.2 Frequency Dependence of $T_g$ :

### Vogel-Fulcher Law:

Almost all types of glasses (including spin glasses) obey Vogel-Fulcher law [8]

$$\tau = \tau_0 \exp \left[ \frac{A}{T_g(\omega) - T_0} \right] \quad (6.6)$$

rather than Arrhenius law

$$\tau = \tau_0 \exp \left[ \frac{A}{T} \right]$$

where  $\tau_0$  is independent of the frequency of measurement, although  $T_g$  depends on it crucially. The latter dependence will be studied in this section. Since  $\tau$  diverges at  $T_g(\omega) = T_0$ ,  $T_0$  is a true transition point revealed by a static measurement. Hence, we identify  $T_0$  with  $T_g$  in section 6.1. Thus all cooperative effects will manifest themselves via  $T_0$  (i.e.,  $T_g$ ).

We now derive (6.6) from the basic model developed in section 6.1. Let us concentrate on the cluster, say  $i$ th, that grows to infinity at  $T_g$ . Hence the number of spins in the  $i$ th cluster should be given by

$$n_i \sim (T - T_g)^{-\gamma}$$

which diverges at  $T = T_g$ , as required. On the other hand, the life time of this cluster can be written as

$$\tau = \tau_0 \exp ( |F_i| / k_B T )$$

where  $F_i$  is the free energy of this  $i$ th cluster, i.e.,  $F_i$  is the free energy needed to overcome the barrier.

Since the cluster is in thermal equilibrium with the free spins at any temperature the free energy per spin within the cluster is  $k_B T \ln (2S+1)$ , so that

$$\tau = \tau_0 \exp [n_i \ln (2S+1)] \quad (6.7)$$

i.e.,

$$\tau = \tau_0 \exp \left[ \frac{A}{(T - T_g)^\gamma} \right]$$

where,

$$A = (\text{const}) \ln(2S+1).$$

For a.c. susceptibility measurement, the cluster will appear frozen as soon as  $\tau > \frac{1}{\nu_m}$  where  $\nu_m$  is the frequency of measurement. Thus

$$\nu_m = \nu_0 \exp \left[ - \frac{A}{\{T_g(\omega) - T_g(0)\}^\gamma} \right] \quad (6.8)$$

We call (6.8) Vogel Fulcher type law; for Vogel-Fulcher law  $\gamma = 1$ . From experimental measurements Tholence [301] found  $\gamma = 1$ , although he maintained that more high-frequency measurements are required to either confirm this or obtain a more reliable value of  $\gamma$ . For Bethe lattice  $\gamma = 1$  [353]. Chalupa [357], on the other hand, found  $\gamma = 15$ . However, his ideas were different from ours. He maintained that the number of clusters is  $\Omega / \xi^3$  ( $\xi$  = correlation length) or that clusters have a characteristic volume. This is not true in our percolative picture. Cluster sizes in our picture grow inhomogeneously and we have a distribution of



cluster sizes and their corresponding relaxation times. This fact will be utilized in a later chapter.

Shtrikman and Wohlfarth [ 296 ] derived Vogel-Fulcher law within their model of blocking of superparamagnetic clusters. In the strong coupling regime, i.e., when inter-cluster interaction is not negligible, Shtrikman and Wohlfarth had to assume temperature-dependent effective cluster size. This idea seems to be closer to ours. The relation between Shtrikman-Wohlfarth approach and ours will become clear in the next chapter.

### 6.3 q-Dependence of $T_g$

Neutron scattering data in spin glasses were analyzed by Murani [ 37,85 ] to show that  $\chi(q)$  exhibits a maximum at  $T_g(q)$  which decreases monotonously with increasing  $q$ -value. Bhargava and Kumar's [358] treatment based on EA model yield  $q$ -independent  $T_g$ . Levin et.al.'s cluster model [ 220-222 ] also yields a  $q$ -independent  $T_g$ . The latter work has been severely criticized by Murani [223] (see section 1.4.2 of Chapter 1 for a discussion). On the other hand, Murani [ 37,85 ] argued that at finite  $q$  the characteristic length scale probed is  $\sim \frac{1}{q}$  and hence only clusters of size  $\geq 1/q$  will appear frozen. At  $T_g$ , where an infinite cluster first forms, there are still a finite fraction of spins in clusters of size  $1/q$  which have lifetimes smaller than the measurement time  $\tau_m$ . Only at lower temperatures all these clusters join the infinite cluster in being frozen. This would lead to a  $T_g(q)$  lower than

$T_g (q=0)$ . In this section we shall quantify Murani's ideas using the standard scaling laws of percolation theory [359].

Let  $C(q)$  be the correlation function of our percolation theory. This is directly related to the 'pair connectedness' function of Essam [359].  $C(\underline{r}-\underline{r}', p)$  is defined as the joint probability of

- (a) the point  $\underline{r}$  being occupied by a sphere centre which belongs to a finite cluster and
- (b) the point  $\underline{r}'$  also being occupied by a sphere centre which belongs to the same cluster.

$p(T)$  is the volume fraction. If  $P(p)$  be the percolation probability i.e., the probability of a sphere joining an infinite cluster, then the probability of the first event mentioned above is  $p_a = p [1 - P(p)]$ . Moreover, defining  $\eta(\underline{r} - \underline{r}') = 1$  if  $\underline{r}$  and  $\underline{r}'$  are connected in the sense of (b) and zero otherwise. Then

$$C(\underline{r} - \underline{r}', p) = p_a \langle \eta(\underline{r} - \underline{r}') | a \rangle$$

Also, we have

$$n_1(p) = \sum \langle \eta(\underline{r} - \underline{r}') | a \rangle = \frac{1}{p_a} \sum_{\underline{R}} C(\underline{R}, p)$$

where,

$$\underline{R} = \underline{r} - \underline{r}'.$$

Let us define  $\tilde{C}(q, p)$  as the Fourier transform of  $C(\underline{R}, p)$ ; then

$$p_a n_1 = \tilde{C}(0, p).$$

The life time of a cluster is

$$\tau_{\underline{q}} = \tau_0 \exp \left[ \frac{\tilde{C}(\underline{q}, p) \ln (2S+1)}{p_a} \right] \quad (6.9)$$

as seen by a probe of characteristic length  $1/q$ . As  $q \rightarrow 0$ ,  $\frac{\tilde{C}(\underline{q}, p)}{p_a} \rightarrow n_i$  and (6.9) reduces to (6.7) which was valid for  $q=0$  probes.

Note first that  $p_a$  is a function which does not diverge at any temperature, from the scaling ideas

$$\tilde{C}(\underline{q}, p) = \xi^{2-\eta} \tilde{g}(q \xi),$$

where  $\tilde{g}$  is a universal function for problems in this class, with  $\tilde{g}(0)$  equals constant  $g_0$  and  $g(r) = g_\infty r^{-2+\eta}$  for large values of  $r$ . This gives  $\tilde{C}(0, p) = \xi^{2-\eta} g_0 \propto (T - T_g)^{-\gamma}$ , diverging, as expected, at  $T_g$ ; and,  $\tilde{C}(\underline{q}, p) = g_\infty q^{-2+\eta}$  for  $p = p_c$ . If  $\tau_m$  is the characteristic time of our probe then  $X(\underline{q})$  has a maximum at  $T_g(\underline{q})$  which is a solution of

$$\left[ \frac{\ln (\tau_m / \tau_0)}{\ln (2S+1)} \right] = \frac{\tilde{C}(\underline{q}, p)}{p_a} \quad (6.10)$$

With the weak temperature dependence of  $p_a$ , near  $T_g$  the right-hand side behaves as  $g' q^{-2+\eta}$ , whereas it goes to zero as  $T \rightarrow \infty$ ; in between it is presumably a smoothly decreasing function. The right hand side of equation (6.10) is plotted as a function of  $T$  for various values of  $q$  in Figure 23. From the intercept shown in the graph for the solution of equation (6.10) it is clear that  $T_g(\underline{q})$  decrease with increasing  $q$ , in agreement with experiment.

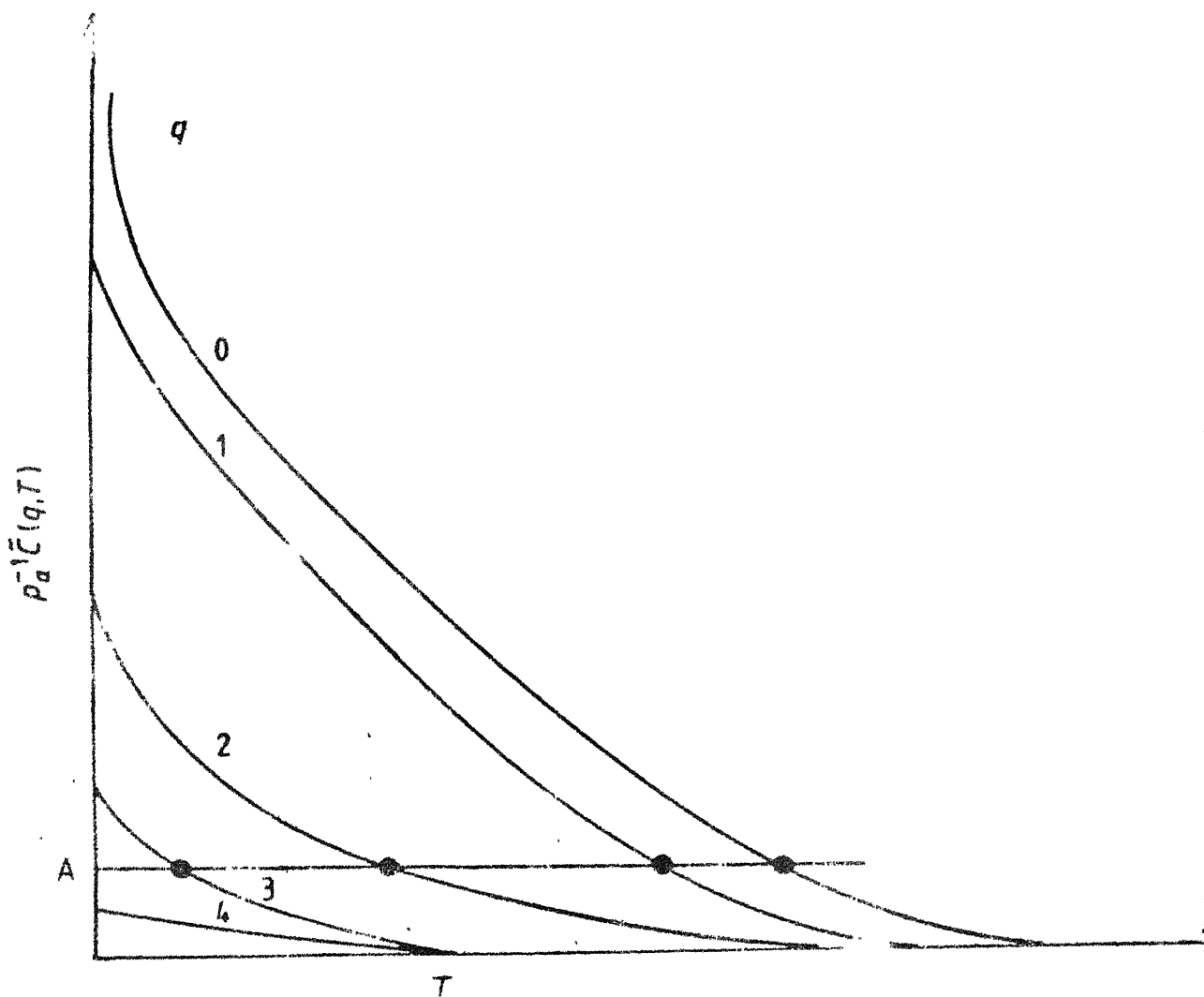


Figure 33 : Schematic plot of  $\tilde{C}(q, T)$  as a function of  $T$  for various values of  $q$ . The intercepts indicate the solutions for  $T_g(q)$ . The intercept of the horizontal line occurs at  $\ln(\tau_M/\tau_0)/\ln(2S+1)$  (point A).

#### 6.4 Linear Response Theory and Zero-Field Low-Frequency AC Susceptibility:

Since zero-field susceptibility is an idealized concept and a finite, although small, external magnetic field is needed to measure susceptibility, we shall use the terms low-field susceptibility and zero-field susceptibility interchangeably.

A well known result of linear response theory (LRT) is

$$\chi(T) = \frac{C}{TNS(S+1)} \sum_{ij} \{ [\langle \underline{S}_i \cdot \underline{S}_j \rangle]_{av} - \lim_{t \rightarrow \infty} [\langle \underline{S}_i(t) \underline{S}_j(0) \rangle]_{av} \} \quad (6.11)$$

which expresses the response function, namely, low frequency a.c. susceptibility  $\chi(T)$  in terms of fluctuations. Here  $C = \frac{(g\mu_B)^2 NS(S+1)}{3k_B}$  is the Curie constant. Neglecting short range order,

$$\sum_{ij} [\langle \underline{S}_i \cdot \underline{S}_j \rangle]_{av} = \sum_{ij} [\langle \underline{S}_i \cdot \underline{S}_j \rangle]_{av} \delta_{ij} = NS(S+1) \quad (6.12)$$

Similarly,

$$\lim_{t \rightarrow \infty} \sum_{ij} [\langle \underline{S}_i(t) \underline{S}_j(0) \rangle]_{av} = \sum_i P(T) Q(T) = NP(T) Q(T) \quad (6.13)$$

where  $P(T)$  = fraction of spins included in the infinite cluster in the thermodynamic limit, and

$$Q = \lim_{t \rightarrow \infty} \frac{1}{NP} \sum_{\substack{i \in \text{inf.} \\ \text{clus.}}} [\langle \underline{S}_i(t) \underline{S}_i(0) \rangle]_{av} \quad (6.14)$$

is the EA order parameter. Notice that while carrying out the spatial averaging in (6.14) we have averaged over the NP sites included in the infinite cluster instead of all the N sites. This is so because only the spins in the infinite cluster have non-zero autocorrelation in the limit  $t \rightarrow \infty$ . Substituting (6.12) and (6.13) into (6.11) we get

$$\chi(T) = \frac{C}{T} \left[ 1 - \frac{P(T) Q(T)}{S(S+1)} \right] \quad (6.15)$$

Similarly, in case of classical spins of unit 'length', i.e.,  $S^2 = 1$ , we get [351]

$$\chi(T) = \frac{C}{T} [ 1 - P(T) Q(T) ] \quad (6.16)$$

It is often argued that the disagreement between Fischer's and Parisi's expressions for  $\chi(T)$  indicates that LRT breaks down in spin glasses because Fischer used relation (6.11), a standard result of LRT. But one should notice that in addition to LRT Fischer also used homogeneous MFT of cooperative phase transition model (CPTM) (i.e.  $P(T) = 1$  for all  $T \leq T_g$ ). In our model LRT holds good although CPTM was assumed to be inadequate [360].

In a finite-sized spin glass, the lifetime of the biggest cluster, even below  $T_g$ , will have a finite (although large) lifetime  $\tau_{\max}$ . Hence, in such cases  $P(T) = 0$  for  $t \gg \tau_{\max}$  and we shall have  $\chi(T) = \frac{C}{T}$  i.e., a Curie-like behaviour. Such behaviour of susceptibility in spin glasses have been observed in computer simulations on finite

systems [24,361] . In other words, spin glass state is not an equilibrium phase in the static sense but only in dynamic equilibrium for  $t \ll \tau_{\max}$ . So we should write (6.16) as

$$\chi_{de}(T) = \frac{C}{T} [1 - P(T) Q(T)] \quad (6.16a)$$

On the other hand, all the spins ( $P=1$ ) appear frozen to any probe of very short characteristic measurement time for all  $T \leq T_g$ . Hence the ideal non-equilibrium susceptibility will be given by

$$\chi_{ne}(T) = \frac{C}{T} [1 - Q(T)] \quad (6.17)$$

which is identical with EA result.

## CHAPTER 7\*

### RELATION BETWEEN PERCOLATION MODEL, EA MODEL AND WOHLFARTH MODEL

In this chapter the mathematical relation between our percolation model and EA model and that between percolation model and Wohlfarth model will be established and interpreted physically [362].

#### 7.1 Relation Between Percolation Model and EA Model

Putting  $P(T) = 1$  for all  $T \leq T_g$  in equations (6.16) and (6.15) we get EA and Fischer's results (1.27a) and (1.30) respectively. This is expected because  $P(T) = 1$  for all  $T \leq T_g$  implies cooperative freezing of all spins at  $T_g$  which is, indeed, the physical picture of the EA cooperative phase transition model

#### 7.2 Relation Between Percolation Model and Wohlfarth Model

In the Wohlfarth model the magnetic susceptibility at temperature  $T$  is given by [287]

$$\chi(T) = \frac{C}{T} \int_0^T f(T') dT' \quad (7.1)$$

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\* Various parts of this chapter have been  
(a) published in J.Phys. F13, L19 (1983)  
(b) submitted for publication.



where  $C$  is the Curie constant and  $f(T)$  is the distribution of blocking temperatures, i.e.  $f(T) \Delta T$  is the fraction of blocking temperatures available in the temperature interval between  $T$  and  $T + \Delta T$ .

The physical meaning of (7.1) is very clear. At temperatures  $T$  only those clusters whose blocking temperatures  $T_B < T$  will contribute to the susceptibility, i.e., those clusters which have not yet 'frozen'. Alternatively, rewriting (7.1) we get

$$\chi(T) = \frac{C}{T} - \frac{C}{T} \int_T^{\infty} f(T') dT' \quad (7.2)$$

since according to the definition,  $f(T)$  is normalized, i.e.,

$$\int_0^{\infty} f(T) dT = 1 \quad (7.3)$$

If all the clusters were unblocked, as is true at high temperatures only,  $\chi(T)$  would be equal to the first term in equation (7.2). However, at lower temperatures the second term, which represents the contribution of blocked clusters, has to be subtracted. The aim in this section is to find the relation between (7.1) and (6.16) .

Let us, for the time being, assume that

$$f(T) = - \frac{d(PQ)}{dT} \quad (7.4)$$

This is the main result of this chapter and will be justified very soon. Substituting (7.4) into (7.1) we get

$$\chi(T) = \frac{C}{T} [P(0) Q(0) - P(T) Q(T)] \quad (7.5)$$

But  $P(T=0) = 1$ , according to definition, and  $Q(T=0) = 1$ , for classical spins of unit length.

Hence (7.5) reduces to

$$X(T) = \frac{C}{T} [1 - P(T) Q(T)]$$

which is identical with (6.16).

Let us now interpret equation (7.4) physically. If there is a large (small) fraction of blocking temperatures  $f(T) \Delta T$  in the temperature range  $T$  to  $T + \Delta T$ , then as one goes from  $T + \Delta T$  to  $T$  the change  $\Delta P$  in the function  $P$  will be large (small) because a large (small) number of spins will coalesce with the infinite cluster. Naturally, the question arises ~~as~~ to why we should take the change in the product  $PQ$ , i.e.  $\Delta(PQ)$  rather than simply  $\Delta P$  in equation (7.4). This is not very surprising. Although nonzero  $P$  'indicates' spin glass ordering, it is insufficient as a 'measure' of the order. Any system of 'particles' interacting via  $\frac{1}{r^3}$  type attractive interaction may undergo a percolation transition of the type considered in Chapter 6. However, such a geometrical transition does not necessarily imply a spin glass type ordering ~~below~~ the percolation threshold. In other words, EA order parameter  $Q$  is a measure of this ordering (some sort of spin stiffness). Therefore, one should take the product function  $PQ$  as a measure of the spin glass ordering. The negative sign in (7.4) accounts for the fact that  $PQ$  increases with decreasing temperature.

For quantum spins  $S$ , (7.4) gets modified to

$$f(T) = - \frac{d}{dT} (PQ/S(S+1)) \quad (7.6)$$

Proceeding as before and remembering that  $Q(T=0) = S(S+1)$  in this case (see Appendix 1) we get from (7.1)

$$\chi(T) = \frac{C}{T} \left[ 1 - \frac{P(T) Q(T)}{S(S+1)} \right]$$

which is identical with (6.15).

The results  $Q(T=0) = 1$  for classical spins of unit 'length' and  $Q(T=0) = S(S+1)$  for quantum spins were derived in appendices within the framework of EA model. But since  $P(T=0) = 1$ , the results of EA treatment and percolation model should be identical at  $T=0$ .

Another similarity between the Wohlfarth model and percolation model is the fact that the former even allows for a temperature-dependent cluster size in order to be consistent with the observed Vogel-Fulcher law [296], though it does not bring percolation into the picture.

It is very interesting to note that an equation exactly similar to (7.4) has been derived by Murani for the distribution of relaxation time  $\tau$  as

$$g(\tau) = - \frac{d \xi(\tau)}{d \tau} \quad (7.7)$$

where  $\xi(\tau)$  is the time-correlation function in the limit momentum transfer  $q \rightarrow 0$ , and  $g(\tau)$  is the distribution of relaxation times. Since there is a one-to-one correspondence

between blocking temperature and relaxation times, we think that equation (7.7) supports our view. Moreover,  $\chi(\tau)$  is related to  $g(\tau)$  by the equation

$$\chi(\tau) = \frac{C'}{T} \int_0^{\tau} g(\tau') d\tau' \quad (7.8)$$

which is formally similar to equation (7.1) and can be similarly interpreted. Since only clusters with relaxation times  $< \tau$  contribute to  $\chi(\tau)$  the upper limit of the integral on the R.H.S. of (7.8) is  $\tau$ .

It appears at first sight that since even finite clusters can freeze in Wohlfarth model it will invalidate the derivation of equation (6.15) and (6.16) from (7.1). But it is not so. Finite cluster freezing implies  $P(T) \neq 0$  even for  $T > T_g$ . But since  $P(T_g)$  has not been utilized in the above derivation our treatment remains valid. The only modification necessary for finite cluster freezing is to call  $P(T)$  in equations (6.15) and (6.16) the 'fraction of spins blocked' instead of 'fraction of spins belonging to the infinite cluster'. We summarize this section saying that it is the percolating spin-clusters that mimic the 'grains fins' of the Wohlfarth model so convincingly. But unlike the Wohlfarth model, spins belonging to different clusters interact via RKKY interaction as described in Chapter 6.

### 7.3 Calculation of $f(T)$ [363]

Although Wohlfarth model does not provide any prescription for abinitio calculation of  $f(T)$ , the latter can be obtained from experimentally measured values of  $\chi(T)$  using relation (7.1), i.e.,

$$f(T) = \frac{d(T\chi)}{dT} / \lim_{T \rightarrow \infty} (T\chi) \quad (7.1a)$$

Van Duynveldt and Mulder [288] calculated  $f(T)$  from their susceptibility data using relation (7.1a). Surprisingly,  $f(T)$  thus calculated exhibits a sharp peak, near  $T_g$ , the temperature corresponding to the susceptibility maximum. From this observation Shtrikman and Wohlfarth [296] concluded that such apparently cooperative effect around  $T_g$  arises due to inter-cluster interaction. It is difficult to account for the observed behaviour of  $f(T)$  within the original framework of non-interacting clusters of the Wohlfarth model. In this section we calculate  $f(T)$  by an alternative method comparing the experimental data on susceptibility with our theory (i.e., using equations (6.16a), (6.17) and (7.4)). As a result we gain an insight into the origin of the cooperative effects in the gradual freezing of spin clusters. This cooperative effect arises from the intercluster interaction which is taken into account in the percolation model.

We can rewrite equation (6.16a) as

$$\chi_{de} = \frac{C}{T} (1 - Q + \Delta) \quad (7.8)$$

(which apparently looks similar to Sompolinsky's result [117])

and

$$\Delta = (1 - P) Q \quad (7.9)$$

On the other hand

$$x_{ne} = \frac{C}{T} (1 - Q) \quad (7.10)$$

Yeshurun and Sompolinsky (YS) [364] found the same expressions (7.8) and (7.10) for  $x_{de}$  and  $x_{ne}$  although the microscopic description (i.e., Sompolinsky's model [117]) underlying their derivation is different from ours. However, this difference does not affect our reasoning here because neither YS nor we calculate  $\Delta$  and  $Q$  from microscopic considerations. Using equations (7.8) and (7.10) and the well known fact [76] that a field-cooled spin glass achieves its equilibrium state, YS obtained  $\Delta$  and  $Q$  from their experimental data. From these  $\Delta$  and  $Q$  we calculate  $PQ$  (shown in Figure 24) using relation (7.9), i.e.  $PQ = Q - \Delta$ . Using  $PQ$  thus obtained, we calculate  $f(T)$  from equation (7.4).  $f(T)$  plotted in Figure 25 is in remarkably good qualitative agreement with that obtained by VM [288]. From  $PQ$  and  $Q$  values we calculated  $P$  as shown in Figure 24. It is typical of any percolation probability: the probability rises very sharply near the percolation threshold and then flattens off.

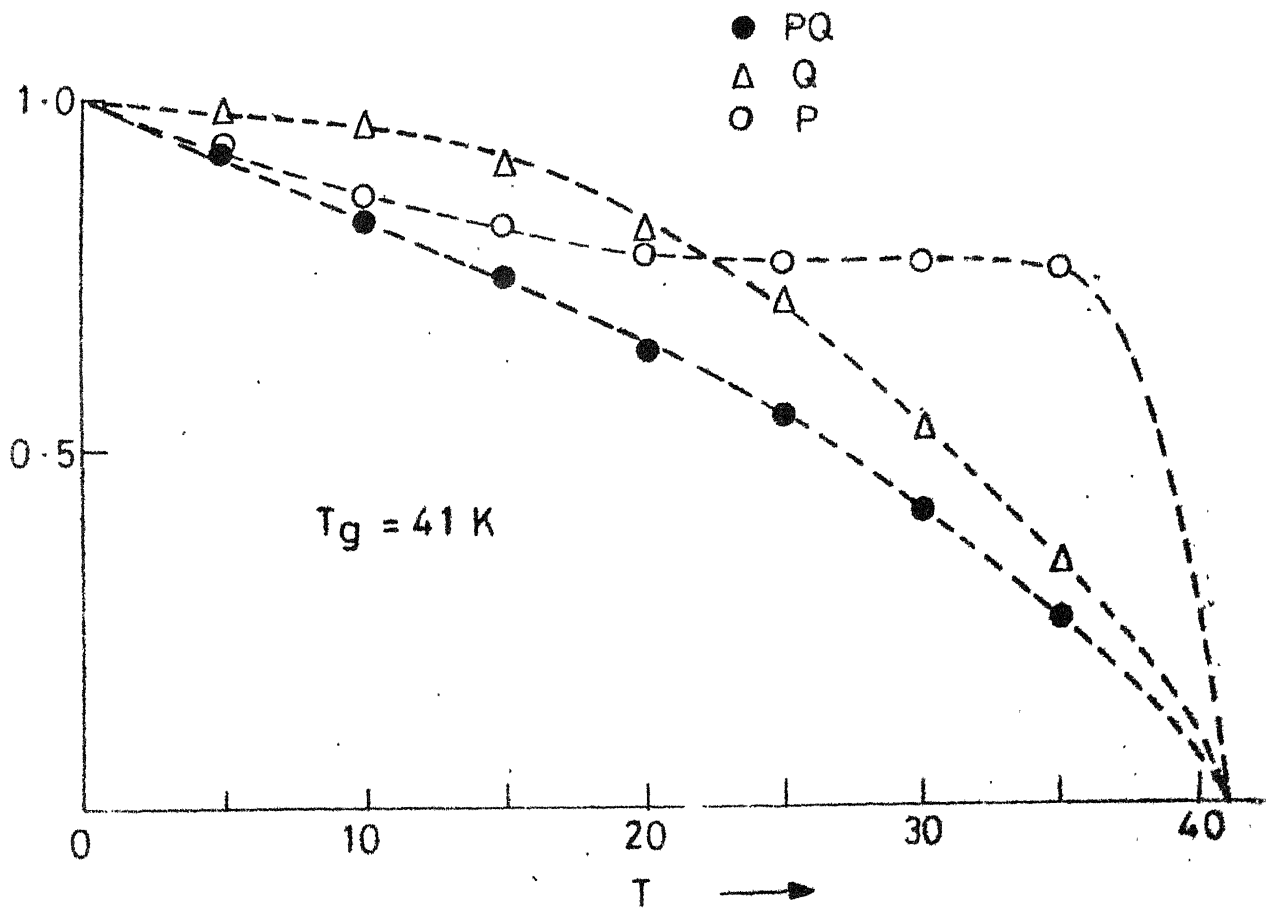


Figure 24 : P, PQ and Q shown as a function of temperature for amorphous SG  $(\text{Fe}_{0.64} \text{Mn}_{0.36})_{75} \text{P}_{16} \text{B}_6 \text{Al}_3$ .

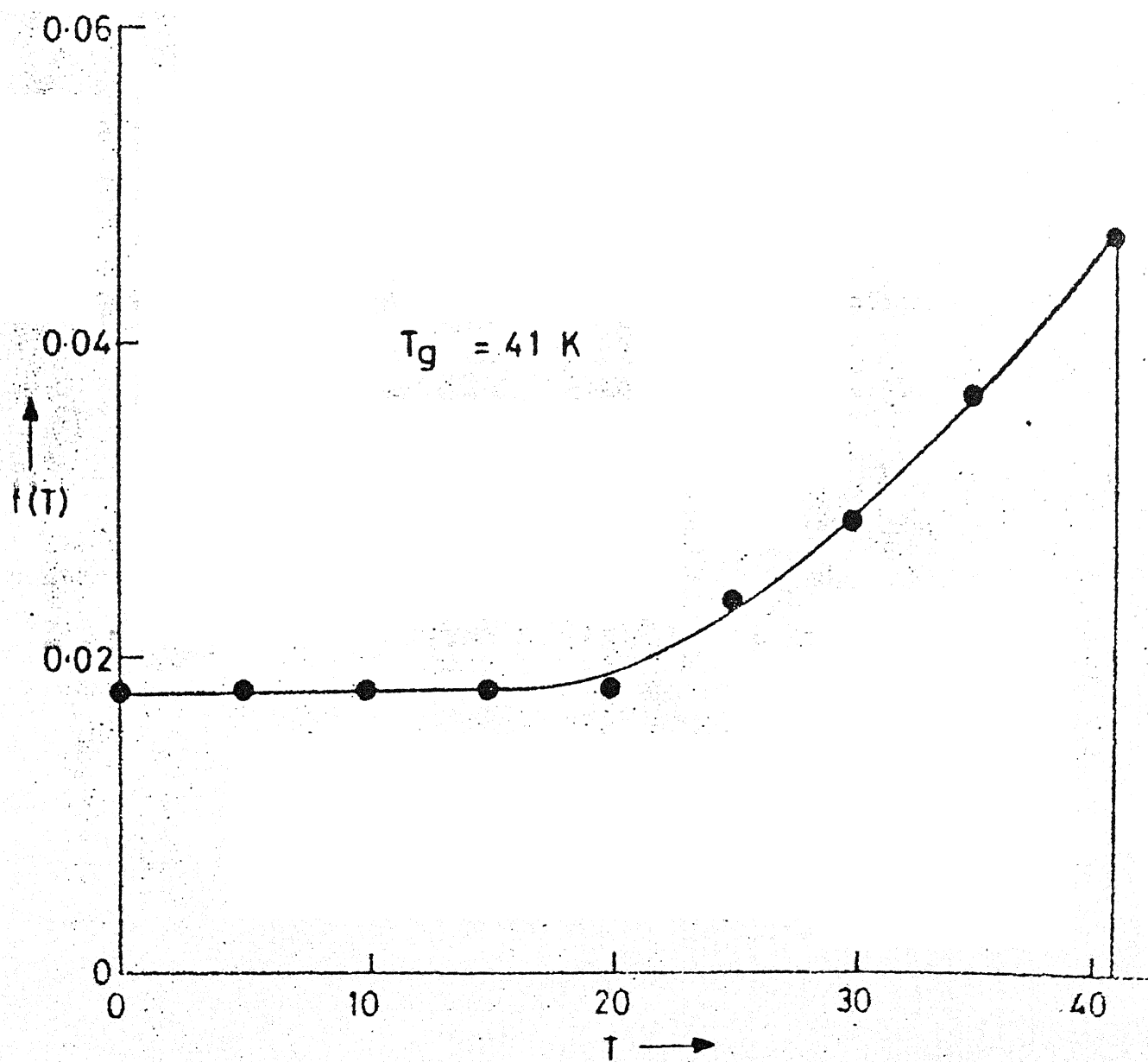


Figure 25 : Distribution of blocking temperatures  $f(T)$  for amorphous SG  $(\text{Fe}_{0.64}\text{Mn}_{0.36})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$ .



## CHAPTER 8\*

### SLOW RELAXATION OF MAGNETIZATION IN SPIN GLASSES AND $1/f$ NOISE

Our aim in this chapter is to derive the logarithmic relaxation (initially exponential for very short time) of magnetization analytically within the framework of the percolation model of spin glass transition developed in the preceding chapters [365-368]. It reveals a deeper connection with  $1/f$  noise in a wide variety of other systems [366].

#### 8.1 Relaxation in the Absence of External Magnetic Field

Logarithmic decay was derived earlier in other similar contexts by Neel [369], Street and Woolley [370] and Gaunt [371]. In case of rock magnetism Neel [369] assumed the existence of a distribution of the volume of magnetic 'particles' and coercive force while Street and Wooley [370] assumed a distribution of barrier heights. Both these approaches are, of course, physically equivalent. A distribution of relaxation

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iii) accepted for publication in Phys.Lett.A (in press).

times appears very naturally also in our percolation model - it is associated with the distribution of cluster size.

The distribution  $P(n)$  of cluster size  $n$ , i.e., the number of finite clusters containing  $n$  spins can be written as

$$P(n) = N_0 \frac{\exp(-\lambda n)}{n^\mu} \quad (8.1)$$

where  $N_0$  is a normalization constant,  $\lambda$  depends on temperature and  $\mu$  is another positive constant [365]. Value of  $\mu$  for various lattices are well known [372, 373],  $\lambda \propto (T - T_g)^2$ .

Then, for normalization

$$\int_1^{n_{\max}} n P(n) = N_0 \int_1^{n_{\max}} \frac{\exp(-\lambda n)}{n^{(\mu-1)}} dn = N(1-P) = N\Psi, \text{ say,} \quad (8.2)$$

where  $n_{\max}$  is the size of the biggest finite cluster and  $N$  is the total number of spins in the system. Putting  $\lambda n = s$ , (8.2) reduces to

$$N_0 \lambda^{(\mu-2)} \int_{\lambda}^{n_{\max}} e^{-s} s^{(1-\mu)} ds = N\Psi$$

and hence

$$N_0 = N\Psi / \left[ \lambda^{(\mu-2)} \left\{ \lambda^{-\frac{(\mu-1)}{2}} e^{-\lambda/2} W_{\frac{(1-\mu)}{2}, \frac{(2-\mu)}{2}}(\lambda) - (\lambda n_{\max})^{-(\mu-1)/2} e^{-\lambda/2} W_{(1-\mu)/2, (2-\mu)/2}(\lambda n_{\max}) \right\} \right]$$

where  $W_{\nu, \pi}(\lambda)$  is the Whittaker function, the functional argument being  $\lambda$ . Since  $n_{\max}$  is usually very large, replacing  $n_{\max}$  by infinity introduces negligibly small error. Then

$$N_0 = N^{\lambda} \int \lambda^{\frac{(\mu-3)}{2}} e^{-\lambda/2} W\left(\frac{(1-\mu)}{2}, \frac{(2-\mu)}{2}(\lambda)\right) d\lambda \quad (8.3)$$

Let  $M(0)$  be the initial magnetization per site and  $\tau(n)$  be the relaxation time for magnetization per site in a cluster of size  $n$ .  $\tau(n)$  depends on the cluster size  $n$  and all sites in the same cluster have same  $\tau$ , because when a cluster relaxes, all the spins belonging to it relax together. In other words, relaxation time of magnetization per site in a cluster is identical with that of the cluster. The relaxation time of a cluster of size  $n$ , derived in Chapter 6 (equation 6.7), is given by

$$\tau(n) = \tau_0 \exp(nK) \quad (8.4)$$

where,

$$K = \ln(2S + 1)$$

Because of the relation (8.4) between  $n$  and  $\tau$ , a distribution  $P(n)$  of cluster size will always be equivalent to a distribution  $g(\tau)$  of relaxation times.

The magnetization per spin observed at time  $t$

$$\begin{aligned} &= \frac{1}{N} \sum_i \text{magnetization of the } i\text{th spin at time } t \\ &= \frac{1}{N} \sum_{n=1}^{n_{\max}} (\text{magnetization per spin at time } t \text{ in a cluster of size } n) nP(n) \\ &\quad + M_{\infty} \\ &= \frac{1}{N} \sum_{n=1}^{n_{\max}} M(0) \exp[-t/\tau(n)] nP(n) + M_{\infty} \end{aligned}$$

where  $M_{\infty} = \frac{1}{N} \sum_{i \in \text{infinite cluster}} M_i = M(0) P$

because the lifetime of the infinite cluster is infinitely large.

Therefore,

$$\begin{aligned} M(t) &= \frac{M(0)}{N} \int_1^{n_{\max}} n P(n) \exp[-t/\tau(n)] \, dn + M(0) P \\ &= M(0) N'_0 \int_1^{n_{\max}} dn \frac{\exp(-\lambda n)}{n^{(\mu-1)}} \left\{ 1 + \sum_{p=1}^{\infty} \frac{(-\alpha t)^p}{p!} e^{-pnK} \right\} \\ &\quad + M(0) P \end{aligned}$$

where  $N'_0 = N_0/N$  and  $\alpha = \tau_0^{-1}$

Using (8.2),

$$M(t) = M(0) + I$$

where,

$$\begin{aligned} I &= M(0) N'_0 \sum_{p=1}^{\infty} \frac{(-\alpha t)^p}{p!} \int_1^{n_{\max}} dn \frac{\exp[-(\lambda + pK)n]}{n^{(\mu-1)}} \\ &= M(0) N'_0 \sum_{p=1}^{\infty} \frac{(-\alpha t)^p}{p!} (\lambda + pK)^{(\mu-2)} \frac{(\lambda + pK)n_{\max}}{\int du} \frac{e^{-u}}{u^{(\mu-1)}} \end{aligned}$$

where,

$$u = (\lambda + pK)n$$

Hence,

$$I = M(0) N'_0 \sum_{p=1}^{\infty} \frac{(-\alpha t)^p}{p!} (\lambda + pK)^{(\mu-2)}$$

$$\begin{aligned}
& \left[ (\lambda + pK)^{-\frac{(\mu-1)}{2}} e^{-\frac{(\lambda + pK)}{2}} W_{\frac{(1-\mu)}{2}, \frac{(2-\mu)}{2}}(\lambda + pK) \right. \\
& \left. - \{(\lambda + pK)n_{\max}\}^{-\frac{(\mu-1)}{2}} e^{-\frac{(\lambda + pK)}{2}} n_{\max} W_{\frac{(1-\mu)}{2}, \frac{(2-\mu)}{2}}((\lambda + pK)n_{\max}) \right]
\end{aligned}$$

Since  $n_{\max}$  is usually very large we can, as before, drop the second term within the brackets. Using the asymptotic form of the Whittaker function for large  $(\lambda + pK)$  we get

$$I \approx M(0) N'_0 \sum_{p=1}^{\infty} \frac{(-\alpha t)^p}{p!} (\lambda + pK)^{-1} e^{-(\lambda + pK)}$$

We further assume that  $pK \gg \lambda$ , so that

$$\begin{aligned}
I & \approx \frac{M(0) N'_0}{K} \sum_{p=1}^{\infty} \frac{\{-\alpha t / (2S+1)\}^p}{p!} \frac{1}{p} \\
& = \frac{M(0) N'_0}{K} \left[ \text{Ei} \{-\alpha t / (2S+1)\} - 0.577 \right. \\
& \quad \left. - \ln \{\alpha t / (2S+1)\} \right]
\end{aligned}$$

where  $\text{Ei}(z)$  is the exponential-integral function and 0.577 is the Euler's constant. Finally, substituting the series representation of the function  $\text{Ei}(-z)$  we get

$$M(t) = M(0) + A + B \exp\{-\alpha t / (2S+1)\} - S(T) \ln t \quad (8.5)$$

where,

$$A = - \frac{M(0) N_o'}{K} [ 0.577 + \ln \{ \alpha / (2S+1) \} ]$$

and

$$S(T) = \frac{M(0)N_o'}{K}$$

are temperature-dependent but independent of time, and

$$B = \frac{M(0)N_o'}{K} \sum_k (-1)^k \frac{(k-1)!}{\{ \alpha t / (2S+1) \}^k}$$

Therefore, the relaxation is quite fast (almost exponential) initially because of the dominance of the third term in (8.5). However, this third term decays very fast. Therefore, on longer time scales the last term in (8.5) dominates and hence the logarithmic relaxation. For large  $t$  (8.5) reduces to

$$M(t) \simeq M(0) - S(T) \ln t \quad (8.6)$$

In addition to the logarithmic relaxation few more results emerge:

(i) We know that  $\mu < 3$  in general. Then

$$S(T) = \frac{M(0)\lambda^{\frac{(3-\mu)}{2}} e^{\lambda/2\Psi}}{K W_{\frac{(1-\mu)}{2}, \frac{(2-\mu)}{2}}(\lambda)}$$

$S(T) = 0$  at  $T = T_g$  where  $\lambda = 0$ , and  $S(T) \rightarrow \infty$  at  $T=0$  where  $\Psi \rightarrow 0$ . Hence,  $S(T)$  is expected to have a maximum in between  $T=0$  and  $T=T_g$ . Such behaviour of  $S(T)$ , has, indeed, been observed experimentally [76].

- (ii) Since finite clusters exist for  $T > T_g$ , one would expect logarithmic relaxation not only for  $T \leq T_g$ , but also at higher temperatures. Such logarithmic (or almost logarithmic) relaxation even at  $T \approx 6 T_g$  has been observed in monte carlo simulation of canonical spin glasses by Fernandez and Streit [374]. For still higher temperatures the relaxation should be exponential because only single spins (and no bigger cluster) with single relaxation time exist at such high temperatures.
- (iii) One may raise objection against the form (8.1) of  $P(n)$  because it is monotonically decreasing function of  $n$  whereas one would expect  $P(n)$  to exhibit a maximum at an intermediate value of  $n$ . Best fit with computer experiments is obtained with an expression of the form

$$P(n) = N_0 \frac{\exp(d_1 \sqrt{n} - d_2 n)}{n^\mu}$$

where  $d_1 \propto (T - T_g)$  and  $d_2 \propto (T - T_g)^2$  and  $\mu$  is a positive constant. Since at  $T_g$ ,  $P(n) \sim n^{-\mu}$  and  $\sum_n n P(n) = N$ ,  $\mu$  must be at least of the order  $O(1)$ .

For convenience we choose  $\mu = 1$ , so that

$$P(n) = N_0 \frac{\exp(a\lambda' \sqrt{n} - b\lambda' n)}{n} \quad (8.7)$$

where,  $\lambda' = (T - T_g)^2$ , and

$$N_0 = N \Psi \left( \frac{b\lambda'}{a^2} \right) \exp(-a^2/8b) / D_{-2}(-a/\sqrt{2b}),$$

$D_n(z)$  being the parabolic cylinder function. Proceeding exactly as before we again get [367] (8.5). This is not surprising in view of the approximation  $pk \gg \lambda$  adopted in both cases.

- (iv) For magnetic systems Chikazumi [375] considered an example to show how a distribution of relaxation times  $g(\tau)$  leads to slower-than-exponential relaxation.

Chikazumi's distribution

$$g(\tau) = \frac{1}{\ln(\tau_2/\tau_1)} \frac{1}{\tau} \text{ for } \tau_2 > \tau > \tau_1$$

$$= 0 \text{ otherwise} \quad (8.7)$$

can be derived using (8.1) and (8.4) assuming  $\lambda = 0$ ,  $\mu = 1$  and that

$$\ln(\tau/\tau_0) = \ln(\tau_2/\tau_1) = \text{const for } \tau_1 < \tau < \tau_2$$

$$= 0 \text{ otherwise.}$$

- (v) Let us now estimate the time-interval, in terms of  $\tau_0$ , within which (8.6) holds.

Notice that the basic characteristic time scale in the problem is  $2\tau_0$ , because the latter is the relaxation time of a 'loose' single spin in the spin glass. One must remember that  $2\tau_0$  is the renormalized relaxation time of a 'loose' spin (a cluster of size unity) weakly interacting with other spins via damped RKKY interaction rather than that of an isolated 'free' spin. Since the exponential term in (8.5) is negligible for  $t > 2\tau_0$ , equation (8.6) holds for



$$t > t_{\min} = 2\tau_0 \quad (8.8)$$

On the other hand, since we started with positive  $M(0)$ ,  $M(t)$  cannot go negative even after long time. Since the equilibrium value of  $M$  (i.e.,  $M(t = \infty)$ ) is zero, (8.6) can hold only upto a time  $t = t_{\max}$  such that

$$M(0) - M(0) N_0^{-1} K \ln (\alpha t_{\max} / (2s+1)) = 0$$

i.e.,

$$t_{\max} = (2s+1)\tau_0 \exp \left( \frac{1}{N_0 K} \right) \quad (8.9)$$

In other words, (8.6) holds in the time-interval  $t_{\min} < t < t_{\max}$  where  $t_{\min}$  and  $t_{\max}$  are given by (8.8) and (8.9) respectively.

(vi) Notice that no frustration has been explicitly assumed a priori in this calculation. Hence, it follows that frustration is not essential for logarithmic relaxation in spin glasses. Medina et.al.[376] also arrived at the same conclusion from computer simulation.

(vii) We now explore the relation with  $1/f$  noise. It was Kogan [278] who first pointed out that because of the logarithmic relaxation of magnetization, the noise spectrum in spin glasses is proportional to  $1/f$  exactly like  $1/f$  noise in semiconductors [276], dielectric relaxation [277], dislocation mobility [377], etc.

Historically speaking, Schottky, using exponential correlation function (like ours in Chapter 4) with a single relaxation time, obtained a Lorentzian that in the large  $f$  limit yields  $1/f^2$  spectrum rather than  $1/f$  spectrum observed experimentally. Later, a class of models for  $1/f$  noise was proposed based on a distribution of relaxation times. The situation in spin glass physics is clearly similar to this. Besides, Macfarlane [ 378 ] assumed distribution of 'cluster size' which is physically similar to our approach.

- (viii) Although our approach apparently looks similar to that of Stauffer [ 379] there are some fundamental differences. We have considered oscillating RKKY interaction whereas Stauffer considered nearest neighbour ferromagnetic interaction and hence the difference in the definition of 'cluster' in the two models. In Stauffer's model, at very low temperatures, all the spins belonging to the same cluster orient in the same direction whereas in the percolation model, as discussed in Chapter 6, spins get pinned along the spatially random local field and hence may orient along different directions even in the same cluster. Besides percolation in Stauffer's model signals the onset of long range order whereas that in our problem leads only to a random freezing of local magnetization without any long range order in space.

## 8.2 Relaxation in the Presence of External Magnetic Field [368]:

As shown in Chapter 5 mean field approximation with single relaxation time is inadequate to explain the slow relaxation in the presence of external magnetic field. Using the same argument as in section 8.1 we now have

$$M_Z(t) = N'_0 \int [M_Z(eq) + \{M_Z(0) - M_Z(eq)\} e^{-t/\tau(n)}] nP(n) \, dn \quad (8.10)$$

(8.10) yields, for not-too-short time scales,

$$M_Z(t) = [M_Z(eq) + K'] - [K' N'_0 / \ln 2] \ln t \quad (8.11)$$

with  $K' = M_Z(0) - M_Z(eq)$ .

Remembering that it is  $M_Z$  that is measured experimentally, the form (8.11) for magnetization relaxation has been observed by Salamon and Tholence [380].

## CHAPTER 9<sup>\*</sup>

### RESISTIVITY OF SPIN GLASSES IN THE PERCOLATION MODEL

In this Chapter we shall utilize the distribution (8.7) for relaxation times to calculate the resistivity of SG within the framework of the percolation model. Experimental data near  $T_g$  turn out to be in better agreement with the expression to be derived in this Chapter rather than with (4.24). However, for  $T \gg T_g$  (say  $T \gtrsim 6 T_g$ ) the expression (4.24) remains valid though the approximate form (4.25) for  $\phi(T, \tau)$  no longer holds.

In Chapter 4 we derived the following expression for resistivity  $\rho(T)$  with single relaxation time  $\tau$  :

$$\rho(T) = \frac{m^* c N_+ (\epsilon_F)}{n e^2} \left[ V^2 + 3\beta J^2 \left[ \frac{Q(T)}{\beta} + \left\{ \frac{S(S+1)}{3} - Q(T) \right\} \frac{\phi(T, \tau)}{\pi \tau} \right] \right] \quad (9.1)$$

Since in the percolation model only a fraction  $P(T)$  of spins have non-zero long-time ( $t \rightarrow \infty$ ) autocorrelation, equation (9.1) gets modified to

$$\rho(T) = \frac{m^* c N_+ (\epsilon_F)}{n e^2} \left[ V^2 + 3\beta J^2 \left[ \frac{Q(T)P(T)}{\beta} + \frac{S(S+1)}{3} - Q(T)P(T) \right] \int \frac{\phi(T, \tau)}{\pi \tau} g(\tau) d\tau \right] \quad (9.2)$$

where  $g(\tau)$  is given by (8.7). This can also be checked from earlier steps (4.25) - (4.27) where we replaced

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<sup>\*</sup> Submitted for publication.

$\frac{1}{N} \sum_i n_i R_{ZZ}^i(t = \infty)$  by  $cQ$ . The latter replacement was valid in the cooperative phase transition model where all the spins have non-zero autocorrelation for all  $T \leq T_g$ . This is not true in our percolation model as stated in Chapters 6 to 8.

Now,

$$\begin{aligned} & \int \frac{\phi(T, \tau)}{\pi \tau} g(\tau) d\tau \\ &= \frac{1}{\ln(\tau_2 / \tau_1)} \int_{\tau_1}^{\tau_2} \frac{\pi k_B^2 T^2 \tau^2}{(\pi k_B T \tau + 3)} \left( \frac{1}{\tau^2} \right) d\tau \\ &= \frac{\ln[(\pi k_B T \tau_2 + 3) / (\pi k_B T \tau_1 + 3)]}{\beta \ln(\tau_2 / \tau_1)} \end{aligned} \quad (9.3)$$

Substituting (9.3) into (9.2) we get

$$\begin{aligned} \rho(T) = \frac{m^* c N_+ (\epsilon_F)}{ne^2} \left[ V^2 + 3 J^2 \left[ QP + \left\{ \frac{S(S+1)}{3} - QP \right\} \right. \right. \\ \left. \left. \frac{\ln[(\pi k_B T \tau_2 + 3) / (\pi k_B T \tau_1 + 3)]}{\ln(\tau_2 / \tau_1)} \right] \right] \quad (9.4) \end{aligned}$$

Notice that since  $Q(0)P(0) = S^2/3$  and  $\ln 1 = 0$ ,

$$\rho(0) = \frac{m^* c N_+ (\epsilon_F)}{ne^2} [V^2 + J^2 S^2]$$

and,

$$\rho(\infty) = \frac{m^* c N_+ (\epsilon_F)}{ne^2} [V^2 + J^2 S(S+1)]$$

which are the correct Yoshida limits.

This is rather fortuitous, because the distribution (8.7)

and hence (9.5) holds only near  $T_g$ . Rewriting equation (9.4)

as,

$$\begin{aligned} \rho(T) = & \frac{m^* c N_+(\epsilon_F)}{n e^2} \left[ V^2 + J^2 S(S+1) \right. \\ & + 3J^2_{QP} \left[ 1 - \frac{\ln(3 + \pi k_B T \tau_2)/(3 + \pi k_B T \tau_1)}{\ln(\tau_2/\tau_1)} \right] \\ & \left. - J^2 S(S+1) \left[ 1 - \frac{\ln(3 + \pi k_B T \tau_2)/(3 + \pi k_B T \tau_1)}{\ln(\tau_2/\tau_1)} \right] \right], \end{aligned}$$

$$\rho(T_g) = \rho(\infty) - \langle H \rangle$$

where,

$$\langle H \rangle = J^2 S(S+1) \left[ 1 - \frac{\ln\{(3 + \pi k_B T \tau_2)/(3 + \pi k_B T \tau_1)\}}{\ln(\tau_2/\tau_1)} \right] > 0,$$

and hence  $\rho(T_g) < \rho(\infty)$ .

Next we calculate the slope of  $\rho(T)$  curve. From

(9.4)

$$\begin{aligned} \left( \frac{n e^2}{m^*} \right) \frac{1}{3J^2 c N_+(\epsilon_F)} \frac{d\rho}{dT} = & \frac{d(PQ)}{dT} \left[ 1 - \frac{\ln\{(3 + \pi k_B T \tau_2)/(3 + \pi k_B T \tau_1)\}}{\ln(\tau_2/\tau_1)} \right] \\ & + \frac{1}{\ln(\tau_2/\tau_1)} \left\{ \frac{S(S+1)}{3} - PQ \right\} \left[ \frac{\pi k_B \tau_2}{3 + \pi k_B T \tau_2} - \frac{\pi k_B \tau_1}{3 + \pi k_B T \tau_1} \right] \end{aligned} \quad (9.5)$$

We can rewrite equation (9.5) as,

$$\begin{aligned} \left( \frac{n e^2}{m^*} \right) \frac{\ln(\tau_2/\tau_1)}{3J^2 c N_+(\epsilon_F)} \frac{d\rho}{dT} = & \frac{d(PQ)}{dT} \left[ \ln \left( 1 + \frac{3}{\pi k_B T \tau_1} \right) - \ln \left( 1 + \frac{3}{\pi k_B T \tau_2} \right) \right. \\ & \left. + \left\{ \frac{S(S+1)}{3} - PQ \right\} \left[ \frac{3 \pi k_B (\tau_2 - \tau_1)}{(3 + \pi k_B T \tau_1)(3 + \pi k_B T \tau_2)} \right] \right] \end{aligned} \quad (9.6)$$

First of all, notice that the factors within the brackets [ ] in the equation (9.6) are positive because  $\tau_2 \gg \tau_1$ . Next slightly recasting equation (9.6) we get

$$\begin{aligned} \left(\frac{ne^2}{m^*}\right) \frac{\ln(\tau_2/\tau_1)}{3J^2 cN_+(\epsilon_F)} \frac{d\rho}{dT} = \frac{d(PQ)}{dT} \left[ \ln\left(\frac{\pi k_B \tau_2}{3 + \pi k_B T \tau_2}\right) \right. \\ \left. - \ln\left(\frac{\pi k_B \tau_1}{3 + \pi k_B T \tau_1}\right) \right] \\ + \left\{ \frac{S(S+1)}{3} - PQ \right\} \left[ \frac{\pi k_B \tau_2}{3 + \pi k_B T \tau_2} - \frac{\pi k_B \tau_1}{3 + \pi k_B T \tau_1} \right] \end{aligned} \quad (9.7)$$

Since  $\frac{d(PQ)}{dT} < 0$ , and  $\frac{S(S+1)}{3} \gg PQ$ , the first term is negative whereas the second term is positive. Moreover,  $-\frac{d(PQ)}{dT} \ll 1$  as shown in Chapter 7. Remembering that  $\tau_2 \gg \tau_1$ , it is straightforward to show that the factor within square bracket [ ] in the first term is much less than that in the second term. As a result the second term in (9.7) (which is positive)  $\gg$  the first term (which is negative), and hence  $\frac{d\rho}{dT} > 0$ . Notice that  $\frac{d\rho}{dT}$  does not exhibit any maximum at any finite temperature, because at all temperatures  $T < T_g$  the second term is much larger than the first and above  $T_g$  first term vanishes. Since  $\frac{d\rho}{dT} > 0$  even at  $T_g$ , there is no anomaly in  $\rho(T)$  at  $T_g$ , as observed experimentally.

So, we conclude that for  $T$  near  $T_g$ ,  $\rho(T)$  is given by (9.4) where the approximate form (4.25) for  $\phi(T, \tau)$  is valid. On the other hand, for  $T \gg T_g$  (say for  $T \geq 6 T_g$ ) (4.24) holds for  $\rho(T)$  although (4.25) can no longer be used

for  $\phi(T, \tau)$  as shown in Chapter 4. This is so because for  $T \gtrsim 6 T_g$  only single spins (and no bigger cluster) exist. Strongest evidence for cluster formation below  $\sim 6 T_g$  comes from the deviation of  $\chi(T)$  from Curie-Weiss behaviour.

Experimental data on  $\rho(T)$  show a maximum at a finite temperature  $T_m > T_g$  as stated in Chapter 1. This does not follow from our model because of two reasons — firstly our distribution (8.7) which is valid near  $T_g$  may not be applicable near  $T_m$ , and secondly, our treatment goes only upto the order  $J^2$ . The decrease of  $\rho(T)$  beyond  $T_m$  is "reminiscent of the Kondo effect" [343] for which one has to make an analysis upto at least the order  $J^3$  and beyond.



## CHAPTER 10<sup>\*</sup>

### RANDOM WALK AND MAGNETIZATION DISTRIBUTION OF RKKY SPIN CLUSTERS

By RKKY spin clusters we mean clusters of spins interacting via RKKY interaction as defined in Chapter 6. In this chapter we shall apply the theory of random walk to calculate the probability distribution of the resultant magnetization of percolating spin clusters. This method also encompasses quantum spins, a generalization never attempted before.

The basic problem of an unbiased one dimensional random walk can be formulated as follows: "a particle suffers displacements along a straight line in the form of a series of steps of equal length, each step being taken either in the forward or in the backward direction with equal probability  $\frac{1}{2}$ . After taking  $N$  such steps, the particle could be at any of the points  $-N, -N+1, \dots, N-1, N$ . We ask: what is the probability  $W(M, N)$  that the particle arrives at the point a distance  $M$  steps away from the starting point after suffering  $N$  displacements?"[382]. The solution of this problem is well known from the consideration of combinatorics:

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<sup>\*</sup>Submitted for publication.

$$W(M,N) = \frac{N!}{\left[\frac{1}{2}(N+M)\right]!\left[\frac{1}{2}(N-M)\right]!} \left(\frac{1}{2}\right)^N \quad (10.1)$$

which, in the limit of large  $N$  and  $M \ll N$  reduces to

$$W(M,N) = (2/\pi N)^{1/2} \exp(-M^2/2N) \quad (10.2)$$

Rivier and Taylor [383]

remarked that 'spin glasses are realisations of a random walk which is the simplest of random processes. They, therefore, serve as a model for the study of random magnetic matter in general'. To our knowledge the idea of random walk has not been fully utilized for the study of spin glasses. This chapter is an attempt, albeit approximate, in that direction.

### 10.1 Ising Spins

Let us first compare a cluster of Ising spins in the absence of an external magnetic field with a one dimensional unbiased random walk.

#### Random Walk

- i) Total number of steps =  $N$
- ii) Each step can be forward (+) or backward (-) with equal probability  $\frac{1}{2}$
- iii) The net displacement can have any of the values  $-N, -N+1, \dots, N-1, N$ .
- iv) We calculate the probability  $W(M,N)$  that the particle arrives at the point  $M$  after suffering  $N$  displacements.

#### Ising Spin Cluster

- i) Total number of spins =  $N$
- ii) Each spin can be either up (+) or down (-)
- iii) The net moment ( $M_z$ ) can have any of the values  $-N, \dots, N-1, N$ .
- iv) We calculate the probability  $P(M,N)$  that the total moment is  $M$  after counting  $N$  spins

However, there is a crucial difference between a random walk and an Ising spin cluster - two successive steps on a random walk are totally uncorrelated, each with probability  $\frac{1}{2}$  for forward and backward move. But in an Ising cluster, though the probability of 'up' and 'down' orientations are each  $\frac{1}{2}$ , the successive spins counted are correlated via  $m_i = \tanh(\beta H_i)$ . In other words, as if the drunkard was not totally drunk. In order to take this correlation properly into account one has to work with conditional probabilities.  $p_\sigma(\underline{R}, \underline{H})$  = the probability that a spin at distance  $\underline{R}$  from the origin has orientation  $\sigma$  when the resulting field at the origin is directed along  $\underline{H}$ , as was originally suggested by Adkins and Rivier [127] (AR) in a related context\*. But exact calculation of  $p_\sigma(\underline{R}, \underline{H})$  involves higher order correlation function and the hierarchy of such correlation functions has to be truncated somewhere judiciously for some practical calculation. Following AR we shall truncate the hierarchy at the very first step by decoupling the joint probability using the approximation

$$(p_+ - p_-)(\underline{R}, \underline{H}) = \text{sign } \underline{H} \cdot m(T) \quad (10.3)$$

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\* AR calculated  $P(H)$  for a macroscopically large system whereas in our problem the spectrum of cluster size ranges from single spin ( $N=1$ ) to very large values.

i.e. equivalently

$$n_+ - n_- = M(T) \quad (10.3)$$

where  $n_+$  and  $n_-$  are the number of up and down spins in a cluster possessing magnetization  $M$ .

From the above consideration it is clear that under the approximation (10.3), one can write, for large values of  $N$

$$P(M, N) = \left(\frac{2}{\pi N}\right)^{\frac{1}{2}} \exp(-M^2/2N) \quad (10.4)$$

This result (10.4) was derived by Abrikosov [372] without invoking the idea of random walk explicitly. Note that in this case the probability distribution for both the total spin as well as the spin projection (magnetization) is governed by (10.4) since the spin dimensionality is one.

## 10.2 Classical Vector Spins

The analogy drawn in the preceeding section can easily be generalized for the case of classical vector spins which may orient in any direction. From the theory of random walk in three dimensions [382] the probability  $W(\underline{r}, N)$  that the position of the particle after  $N$  displacements is  $\underline{r}$  is given by

$$W(\underline{r}, N) = \left(\frac{3}{2\pi N}\right)^{3/2} \exp[-3r^2/2N] \quad (10.5)$$

Hence we must also have

$$P(\underline{M}, N) = \left(\frac{3}{2\pi N}\right)^{3/2} \exp \left[-\frac{3M^2}{2N}\right] \quad (10.6)$$

Because of isotropy in direction this leads to

$$\begin{aligned} P(\underline{M}, N) d^3M &= 4\pi M^2 P(\underline{M}, N) dM \\ &= \left(\frac{2}{\pi}\right)^{\frac{1}{2}} \left(\frac{3}{N}\right)^{3/2} M^2 \exp(-3M^2/2N) dM \end{aligned} \quad (10.7)$$

which is again identical with Abrikosov's corresponding result [372]. Notice that (10.7) gives us the probability of the total spin rather than the spin projection (magnetization). The probability of the magnetization  $M_z$  may also be obtained directly by integrating over  $M_x$  and  $M_y$  to obtain

$$P(M_z, N) dM_z = \left(\frac{3}{2\pi N}\right)^{\frac{1}{2}} \exp \left[-\frac{3M_z^2}{2N}\right] dM_z \quad (10.8)$$

Although the contents of the sections 10.1 and 10.2 are restatements of earlier known results we stress that those results can be obtained very easily once the analogy between the random walk and spin clusters is recognized.

The derivations of (10.2) and (10.6) were very simple because of the applicability of vector addition for coupling of vector spins. This was a direct consequence of the commutation of the various spin components. If it is possible to visualize the random walk as a process where the particle takes small steps  $\delta x$  in small time interval  $\delta t$  such that  $x=N \delta x$  and  $t=N \delta t$  then in the continuum limit ( $\delta x \rightarrow 0$ ,  $\delta t \rightarrow 0$ ) one gets for  $W(x, t, x_0)$  a Fokker-Planck equation.

Similarly, a continuum limit is possible for classical spins and  $P(M,N)$  satisfies the Fokker-Planck equation (10.2) and (10.8) are solutions of such Fokker-Planck equations.

In the next section we shall see how these ideas are to be modified for quantum vector spins.

### 10.3 Quantum Spins

In this section we shall denote the magnetization (and spin) values of individual spins by small letters and those of clusters by capital letters.

The first remarkable difference between the classical and quantum spins is that the latter can have only  $(2s+1)$  orientations, where  $s(s+1)$  is the eigenvalue of  $s^2$ . This leads to two consequences: the ordinary vector addition cannot be applied for coupling of quantum spins and secondly the continuum limit cannot be taken. In other words,  $P(M,N)$  is now a discrete, rather than continuous, function of  $M$ .

We shall first consider integral spins. Since we are interested in the probability distribution of  $S_z = \sum_{i=1}^N s_{iz}$  and these projections obey simple addition law, the problem again reduces to that of effectively one dimensional random walks. However, there is a difference between this case and the one considered in section 10.1. For Ising spins magnetization can change in each step only by  $+1$  or  $-1$  with equal probability  $\frac{1}{2}$ , and hence only nearest neighbour hopping was allowed in the corresponding random walk. In

the case of Heisenberg spin  $s$ , the magnetization can change in each step by any of the values  $-s, -s+1, \dots, s-1, s$ . Therefore, we have to consider a generalized random walk where the 'drunkard' can hop a 'distance'  $-s, -s+1, \dots, s-1, s$  in a single step with equal probability  $1/(2s+1)$ . We now write  $P_s(M, N)$  for the probability that a cluster of  $N$  spins  $s$  has a net moment  $M$ . It satisfies a difference equation

$$P_s(M, N) = p_0 P_s(M, N-1) + p_1 P_s(M-s+1, N-1) \\ + p_2 P_s(M-s+2, N-1) + \dots + p_{2s} P_s(M+s, N-1)$$

which can be rewritten in a more compact notation as

$$P_s(M, N) = \sum_{l=0}^{2s} p_l P_s(M-s+l, N-1) \quad (10.9)$$

where  $p_l$  is the probability of a jump of length  $l$  in one step. For an unbiased random walk  $p_l = 1/(2s+1)$  for all  $l$ . The ordered set  $(M, N)$  forms a two dimensional grid and our aim is to determine  $P_s(M, N)$  on this grid. The obvious boundary condition for all cases is  $P_s(M, 0) = \delta_{M, 0}$ . (10.10) Before embarking to obtain a general solution of (10.9) let us illustrate with a few examples.

#### Example 1:

Let  $s = 1, N = 2$  so that  $p = 1/3$  and  $P(M, 0) = \delta_{M, 0}$ . Besides, from the consideration of normalization we get the sum rule

$$\sum_{M=-S}^S P_s(M, N) = 1. \quad (10.11)$$

Now,

$$\begin{aligned}
 P_1(M, 1) &= \frac{1}{3} [P_1(M-1, 0) + P_1(M, 0) + P_1(M+1, 0)] \\
 &= \frac{1}{3} [\delta_{M-1, 0} + \delta_{M, 0} + \delta_{M+1, 0}] \\
 &= \frac{1}{3} [\delta_{M, 1} + \delta_{M, 0} + \delta_{M, -1}]
 \end{aligned}$$

Hence,  $P_1(1, 1) = P_1(0, 1) = P_1(-1, 1) = \frac{1}{3}$ , and the sum rule (10.11) is satisfied for  $S = 1$ .

Next,

$$\begin{aligned}
 P_1(0, 2) &= \frac{1}{3} [P_1(-1, 1) + P_1(0, 1) + P_1(1, 1)] \\
 &= \frac{1}{3} \left[ \frac{1}{3} + \frac{1}{3} + \frac{1}{3} \right] = \frac{1}{3}
 \end{aligned}$$

$$\begin{aligned}
 P_1(1, 2) &= \frac{1}{3} [P_1(0, 1) + P_1(1, 1) + P_1(2, 1)] \\
 &= \frac{1}{3} \left[ \frac{1}{3} + \frac{1}{3} + 0 \right] = 2/9
 \end{aligned}$$

$$\begin{aligned}
 P_1(-1, 2) &= \frac{1}{3} [P_1(-2, 1) + P_1(-1, 1) + P_1(0, 1)] \\
 &= \frac{1}{3} \left[ 0 + \frac{1}{3} + \frac{1}{3} \right] = 2/9
 \end{aligned}$$

$$\begin{aligned}
 P_1(2, 2) &= \frac{1}{3} [P_1(1, 1) + P_1(2, 1) + P_1(3, 1)] \\
 &= \frac{1}{3} \left[ \frac{1}{3} + 0 + 0 \right] = 1/9
 \end{aligned}$$

$$\begin{aligned}
 P_1(-2, 2) &= \frac{1}{3} [P_1(-3, 1) + P_1(-2, 1) + P_1(-1, 1)] \\
 &= \frac{1}{3} [0 + 0 + \frac{1}{3}] = 1/9.
 \end{aligned}$$

Again the sum rule is satisfied for  $S = 2$ . Notice that (i) the sum rule (10.11) is satisfied for any  $N$ . Thereby preserving normalizations, (ii),  $P_S(M, N)$  is symmetric with respect to the sign of  $M$ .



Example 2:

Let  $s = 2$ ,  $N = 2$ . In this case  $p = 1/5$  and the boundary condition is  $P_2(M, 0) = \delta_{M, 0}$ .

Proceeding exactly as in example 1 we get

$$P_2(0, 2) = 1/5, \quad P_2(1, 2) = P_2(-1, 2) = 4/25,$$

$$P_2(2, 2) = P_2(-2, 2) = 3/25,$$

$$P_2(3, 2) = P_2(-3, 2) = 2/25$$

$$P_2(4, 2) = P_2(-4, 2) = 1/25$$

and the sum rule (10.11) is again satisfied.

Example 3:

Let  $s = 3/2$ ,  $N = 2$ . Hence  $p = 1/4$  and  $P_{3/2}(M, 0) = \delta_{M, 0}$ . Therefore,

$$P_{3/2}(M, 1) = 1/4 [P_{3/2}(M - 3/2, 0) + P_{3/2}(M - \frac{1}{2}, 0)$$

$$+ P_{3/2}(M + \frac{1}{2}, 0) + P_{3/2}(M + 3/2, 0)]$$

$$= \frac{1}{4} [\delta_{M, 3/2} + \delta_{M, \frac{1}{2}} + \delta_{M, -\frac{1}{2}} + \delta_{M, -3/2}]$$

$$P_{3/2}(0, 2) = \frac{1}{4} [P_{3/2}(-\frac{3}{2}, 1) + P_{3/2}(-\frac{1}{2}, 1) + P_{3/2}(\frac{1}{2}, 1) \\ + P_{3/2}(\frac{3}{2}, 1)]$$

$$= \frac{1}{4} [\frac{1}{4} + \frac{1}{4} + \frac{1}{4} + \frac{1}{4}] = \frac{1}{4}$$

$$P_{3/2}(1, 2) = \frac{1}{4} [P_{3/2}(-\frac{1}{2}, 1) + P_{3/2}(\frac{1}{2}, 1) + P_{3/2}(\frac{3}{2}, 1) \\ + P_{3/2}(\frac{5}{2}, 1)]$$

$$= \frac{1}{4} [\frac{1}{4} + \frac{1}{4} + \frac{1}{4} + 0] = \frac{3}{16} = P_{3/2}(-1, 2)$$

$$\begin{aligned}
 P_{3/2}(2,2) &= \frac{1}{4} [P_{3/2}(\frac{1}{2},1) + P_{3/2}(\frac{3}{2},1) + P_{3/2}(\frac{5}{2},1) \\
 &\quad + P_{3/2}(\frac{7}{2},1)] \\
 &= \frac{1}{4} [\frac{1}{4} + \frac{1}{4} + 0 + 0] = 2/16 = P_{3/2}(-2,2)
 \end{aligned}$$

$$\begin{aligned}
 P_{3/2}(3,2) &= \frac{1}{4} [P_{3/2}(3/2,1) + P_{3/2}(5/2,1) + P_{3/2}(7/2,1) \\
 &\quad + P_{3/2}(9/2,1)] \\
 &= \frac{1}{4} [\frac{1}{4} + 0 + 0 + 0] = 1/16 = P_{3/2}(-3,2).
 \end{aligned}$$

$P_s(M,N)$  for any other half integral values of  $s$  can be calculated similarly.

To get a feeling for the variation of  $P_s(M,N)$  with  $M$  as well as  $N$  for given  $s$  we show the values of  $P_1(M,N)$  in a  $N,M$  grid in table 5, and figure 26. Since the probability is drawn out from the centre to the wings with increasing  $N$  symmetrically, the peak value  $P_1(0,N)$  decreases with increasing  $N$  to preserve the sum rule.

TABLE 5

$N \backslash M$	1	2	3	4
-4				1/81
-3			1/27	4/81
-2		1/9	1/9	10/81
-1	1/3	2/9	2/9	16/81
0	1/3	1/3	7/27	19/81
1	1/3	2/9	2/9	16/81
2		1/9	1/9	10/81
3			1/27	4/81
4				1/81

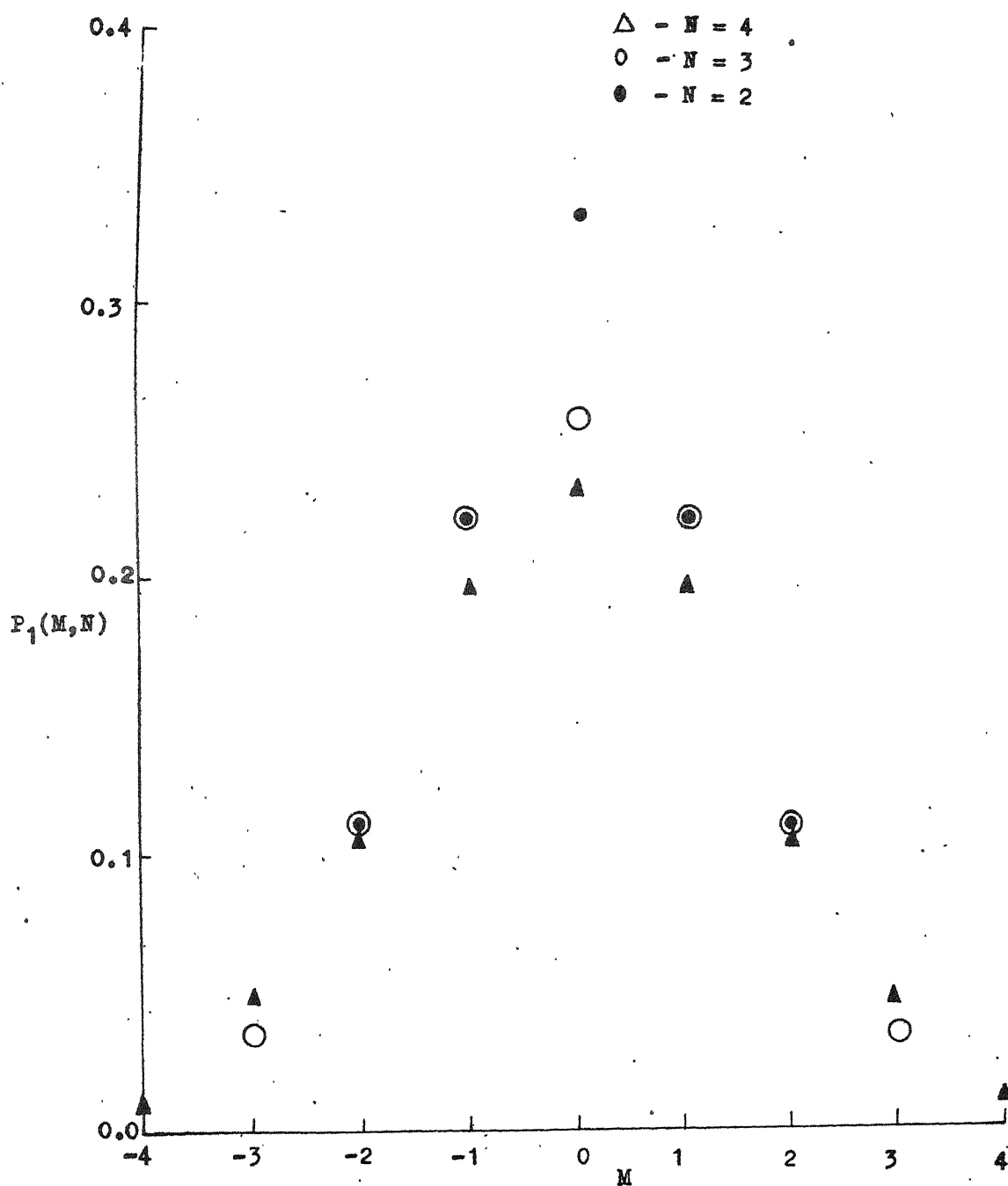


Figure 26 : The probabilities of the magnetizations of clusters of different sizes for spin  $s = 1$ .

#### 10.4 General Solution

In order to solve the general difference equation, let us proceed in two steps. First let us partition the  $N$  steps into a configuration where there are  $n_m$  steps of length  $m$  ( $m = -s, \dots, s$ ). The probability of one such configuration is given by the multinomial expression

$$P_S(\{n_m\}, N) = \frac{N!}{\prod_{m=-s}^s n_m!} (p)^N \quad (10.12)$$

To obtain  $P_S(M, N)$  we can take any configuration which satisfies the conditions

$$\sum_{m=-s}^s n_m = N \quad (10.13a)$$

$$\sum_{m=-s}^s m n_m = M \quad (10.13b)$$

Thus,

$$P_S(M, N) = \sum_{\{n_m\} \in (10.13)} P_S(\{n_m\}, N) \quad (10.14)$$

where the summations to be carried out over all configurations satisfying (10.13). It is straight forward to show that the general solution (10.14) satisfies the difference equation (10.9).

Thus we conclude that the spin (orientational) configuration in a percolating cluster in a RKKY SG can be thought of as the realization of a random walk in some

hyperspace. In addition, the spatial (positional) configuration of the magnetic atoms in a SG can also be thought of as a realization of a random walk. Comparing a RKKY spin cluster with a random ferromagnetic cluster at  $T=0$  we see the following difference: the latter represents an ordered walk (for spin degrees of freedom) on a random walk (for random position of the magnetic atoms in space), whereas the former is the realization of a random walk on a random walk [384]. Rivier and Taylor [383] described a SG as a "drunkard's walk transfixed"; we shall call a SG a "drunkard's walk transfixed on a drunkard's walk".

## CHAPTER 11\*

### 'HOLE' IN THE LOCAL FIELD DISTRIBUTION

We assumed in Chapter 6 that the number of sites where local magnetic field vanishes is vanishingly small at sufficiently low temperatures. In other words, we assumed the existence of a 'hole' in the local field distribution  $P(H)$ . We shall prove it in this chapter.

#### Theorem:

In any spin glass there exists a field  $\delta$  such that  $P(H) = 0$  for  $-\delta < H < \delta$  at sufficiently low temperature.

#### Proof:

To prove this theorem one has to go beyond the simple minded weiss mean field approximation and take the reaction field into account. We stress that one does not necessarily have to work with the TAP model (or, more generally, with any cooperative phase transition model) in order to take the effect of the reaction field into account. The physical origin of the reaction field is much more general and can be derived as follows: the local field produced at the  $j$ th

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\* Submitted for publication.

site by the  $i$ th spin is  $J_{ij}^* M_i$ . The field induces a local moment  $J_{ij} X_j M_i$  at the  $j$ th site, where  $X_j$  is the local susceptibility at the  $j$ th site. This induced moment, again, produces a local field  $J_{ij}^2 X_j M_i$  at the  $i$ th site. Hence the reaction field produced at the  $i$ th site by all other spins  $j (j \neq i)$  is  $\sum_{j \neq i} J_{ij}^2 X_j M_i$ .

If the cavity field at the  $i$ th site (i.e. the local field at the  $i$ th site in the absence of the  $i$ th spin) is  $h_i$ , then

$$h_i = \sum J_{ij} M_j - \sum J_{ij}^2 X_j M_i$$

or

$$H_i = \sum J_{ij} M_j = h_i + \sum J_{ij}^2 X_j M_i \quad (11.1)$$

Notice that so far we have not assumed anything about the nature of the spin glass transition, i.e., whether it is a cooperative phase transition or a gradual freezing process. Let us define the spatial average of a local property  $X_i$  by

$$\bar{X} = \frac{(1/N)}{\sum_i} X_i$$

We may thus replace  $J_{ij}^2 X_j$  by  $A \bar{X}$  (11.2)

where  $A$  is a temperature-independent constant and  $\bar{X}$  is average susceptibility. The right hand side of equation (11.2) is also independent of  $i$  because of statistical homogeneity.

Any homogeneous MFT of CPTM of EA type yields

$$X \propto \beta (1 - Q) \quad (11.3)$$

whereas in the percolation model  $X \propto \beta (1-PQ)$ . From Young and Kirkpatrick's [125] computer simulation and Fischer and Hertz's [389] discussion (so far as the notation is concerned, their  $q$  and  $q$  correspond to our  $Q$  and  $PQ$  respectively), we get

$$1 - Q = \alpha / \beta^2 \quad (11.4)$$

and

$$1 - PQ = \gamma / \beta \quad (11.5)$$

at sufficiently low temperatures;  $\alpha$  and  $\gamma$  being two constants independent of temperature. (11.4) and (11.5) are consistent with each other because  $P < 1$  for all  $T > 0$ .

We also know that

$$M_i = \tanh (\beta h_i) \quad (11.6)$$

because local magnetization is determined by the local cavity field  $h_i$  rather than the total Weiss field  $H_i$ . Therefore as  $T \rightarrow 0$

$$M_i \rightarrow \text{sign} (h_i) \quad (11.7)$$

Substituting (11.5) and (11.7) into (11.1) we get

$$H_i = h_i + \gamma A \text{sign} (h_i) \quad (11.$$

$$\text{or,} \quad |H_i| = |h_i| + |\delta| \quad (11.8)$$

where,

$$\delta = \gamma A$$



The second term on the right hand side of (11.8) is a constant characteristic of the system and does not depend on field. Therefore, the magnitude of the local field i.e.  $|H_1|$  can never be zero; in other words, the probability that  $H_1$  is zero is zero. This proves the theorem.

In any cooperative phase transition mode; since  $\beta(1-Q) = \alpha / \beta$  as  $T \rightarrow 0$  and  $H_1 = h_1$  and there can be no hole in  $P(H)$ . The existence of 'hole' in  $P(H)$  has been observed in computer experiments [315,331].

## CHAPTER 12

### WHERE DO WE STAND?

#### 12.1 What have we Achieved?

##### 12.1.1 Comparison with other Models

Although our percolation model appears similar to that of Smith [353] there are some crucial differences:

- (a) Smith did not take frustration into account. We have shown how  $T_g$  vs.  $c$  curve deviates from linearity because of frustration,
- (b) Smith model is applicable only to Bethe lattice which contains no frustration. Our model is applicable to any realistic three dimensional lattice as well as to amorphous SG,
- (c) Smith suggested, intuitively, that

$$\chi(T) = \frac{C}{T} (1 - P)$$

which differs from our expression (6.16).

In other words, Smith assumes all spins in the infinite cluster to be perfectly stiff (i.e,  $Q = 1$  for all spins in the infinite cluster) whereas we allow for thermal fluctuations and hence incomplete stiffness. Our expression (6.16) has been derived from more fundamental expression (6.11) of LRT.

Our percolation model is qualitatively closer to Cyrot's model [389] because the latter takes frustration into account. However, there are some differences in the quantitative calculations.

- (a) Cyrot, for simplicity, divides the system into "p blocks of n spins where  $np = N$  (N number of spins)". On the other hand, in our percolation model, there is a spectrum of cluster size given by  $P(n)$ .
- (b) In Cyrot's model a SG is distinguished from random ferromagnets as well as from Mattis model by the fact that there is a distribution  $P(F)$ , the probability that a cluster has free energy F. This is so because the number of frustrated bonds varies from cluster to cluster even for same n, and hence the difference in their free energy. In our percolation model, we leave out frustrated sites at the very beginning when we construct spheres of correlation around the spins. Hence, in our model  $F(n)$  is non-random at all temperatures.
- (c) Our derivation of Vogel-Fulcher law is quite different from that of Cyrot [389].

Abrikosov's work [372] is complimentary to ours as stated in Chapters 6 and 10. The relation between Wohlfarth's model and our percolation model has been discussed in Chapter 7. So far as neutron scattering experiments are concerned, our work in Chapter 6 is a quantification of Murani's qualitative ideas [85].

As stated in Chapter 7, our equation (11.8) resembles Sompolinsky's result (1.27d) though the underlying microscopic models are quite different. Sompolinsky's result is valid for only SK model which assumes infinitely weak infinitely long ranged interaction. We derived (11.8) for realistic three dimensional SG with damped RKKY interaction. All the relaxation times diverge in the thermodynamic limit in Sompolinsky's treatment whereas there is a DRT even in the thermodynamic limit in our percolation model. Exactly similarly, our expression (6.16) can be recast into the form (1.27c) where

$$\begin{aligned} Q(x) &= 0 \text{ for } x \leq \bar{x} \\ &= Q_{EA} \text{ for } x > \bar{x}, \text{ with } \bar{x}(T) = 1 - P(T) \end{aligned}$$

But this apparent similarity with Parisi's result (1.27c) does not necessarily imply any deeper connection between our percolation model and SK model.

There seems to be a very close relation between our percolation model and Anderson and coworkers' work [320,321] that describes SG transition as a localization-delocalization problem. Anderson [8] comments that "it is these localized eigenstates which so convincingly mimic the 'clusters' of the Neel interpretation of SG phenomenology". And, we have already shown in Chapter 7 that "it is the percolating spin clusters that mimic the 'grains fins' of the Wohlfarth model so convincingly". Hence we identify the localized eigenstates of the random matrix  $J$  with the finite clusters and

the mobility edge with the percolation threshold. Such a relation was also suspected by Young and Stinchcombe [390]. They also conjectured that the reason why SK model exhibits a phase transition is associated with the corresponding spectrum of  $\mathcal{J}$  which, in this case, does not contain any localized state, which, we think, is true. A rigorous equivalence between our percolation model and localization-delocalization model is under our investigation. This rigorous treatment will make small quantitative changes in our model though the qualitative features are likely to remain unaltered. For example, in equation (6.7)  $\tau_0$  will turn out to be temperature as well as size dependent and the constant factor  $\ln(2S+1)$  will get modified.

One serious objection against Smith model was that clusters become ill-defined for infinite range RKKY interaction. This difficulty can be minimized assuming damped RKKY interaction, as we have done in Chapter 6, because the latter interaction is effectively finite ranged. However, definition of clusters in our model is still ambiguous to some extent. For example, let us consider a cluster of three spins, connected by nearest neighbour ferromagnetic bonds  $J$ , forming a triangle. Such a cluster should freeze when the largest solution of the eigenvalue equation  $\lambda^3 - J^2\lambda - 2J^3 = 0$  is equal to  $k_B T$ , i.e. at a temperature  $T_f = \frac{\lambda_{\max}}{k_B}$ . But defining  $\phi(\lambda) = \lambda^3 - J^2\lambda - 2J^3$  we find that  $\phi(\infty) > 0$  and  $\phi(J) < 0$ , so that  $\phi(\lambda)$  must vanish for a  $\lambda_{\max}$  such that  $J < \lambda_{\max} < \infty$ . In other the cluster under consideration

can freeze at a temperature  $T_f > \frac{J}{k_B}$ , whereas our criterion in Chapter 6 was  $T_f = \frac{J}{k_B}$ . Of course, for low concentration alloys, existence of such clusters as considered above is very unlikely.

The localized state picture makes the definition of clusters exact, and is identical with our definition for highly localized states. For weakly localized states near  $T_g$  (i.e. near the mobility edge) our clusters become fuzzy as large clusters overlap. Hence the description of SG transition as localization-delocalization problem becomes very appropriate. In this picture participation ratio corresponds to the size of the clusters. So far as the dynamics is concerned, calculation of the relaxation time of clusters involves calculation of first passage time.

### 12.1.2 Comparison with Experiments

Our results derived on the basis of percolation model agree well with experimental results:

- (a) Our expression (6.15) and (6.16) exhibit a cusp in  $X(T)$  as observed,
- (b) Our expression (6.8) explains the frequency dependence of  $T_g(\omega)$
- (c) Equation (6.10) explains the  $q$ -dependence of  $T_g(q)$ ,
- (d) Figure 22 agrees with observed  $T_g$  versus  $c$  data,
- (e) Our model accounts for the smooth behaviour of specific heat across  $T_g$ ,

- (f) Figure 25 agrees well with the distribution of blocking temperatures calculated in Ref.[288],
- (g) Logarithmic relaxation of magnetization (8.6) was also derived,
- (h) Equation (9.4) accounts for the smooth behaviour of resistivity  $\rho(T)$
- (i) Equation (5.26) explains the  $M$  dependence of magnetoresistance  $\Delta\rho$  satisfactorily,
- (j) The value of  $J_{sd}$  shown in Table 3 is of the correct order of magnitude,
- (k) Equation (11.8) proves the existence of hole in  $P(H)$  observed in computer experiments,
- (l) In Chapter 6 we also showed that a finite system will become paramagnetic on very long time scales, in agreement with computer simulation results [24,361]

## 12.2 What Else Can be Done?:

As already stated in Section 12.1.1 a rigorous relation between localization-delocalization problem and percolation model is under investigation. This will enable us to clarify some drawbacks of the percolation picture and make more rigorous quantitative predictions which can be compared with experimental data.

We have investigated only two transport properties - resistivity and magnetoresistance. Other properties e.g. thermopower, Hall effect etc. can also be calculated similarly.

None of the percolation models discussed so far examined the effect of spin waves on the stability of the clusters.

Recently SG has been considered as a prototype for many more complicated and less understood problems, e.g. according to Kirkpatrick, "a computer chip is a SG" and according to Hopfield, "the brain is a SG" [391]. Nielsen says, "the universe is a SG" and according to Anderson, "the origin of life is a SG" [391]. These comparisons show the potentiality of spin glass physics. We have shown in this thesis that "a spin glass is a glass" [391]. However, a lot remains to be done. May be, this 'normal science' will end with a "scientific revolution" (in Kuhnian sense)!



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APPENDIX 1Q(T=0) FOR QUANTUM SPINS

It was shown by Fischer [130] that

$$Q = S(S+1) - \frac{1}{(2\pi)^{3/2}} \int d^3r \exp(-\frac{1}{2} r^2) r \zeta^{-\frac{1}{2}} \\ \left[ (S + \frac{1}{2}) \coth (S + \frac{1}{2}) \zeta^{\frac{1}{2}} r - \frac{1}{2} \coth(\frac{1}{2} \zeta^{\frac{1}{2}} r) \right]$$

where  $\zeta = \frac{1}{3} (\beta \Delta)^2 Q$

and  $\Delta$  is the second moment of the gaussian distribution of random bonds,  $P(J_{ij})$ . As  $T \rightarrow 0$ ,  $\beta \rightarrow \infty$  and hence  $\zeta \rightarrow \infty$ . Besides,  $\{(S + \frac{1}{2}) \coth (S + \frac{1}{2}) \zeta^{\frac{1}{2}} r - \frac{1}{2} \coth (\frac{1}{2} \zeta^{\frac{1}{2}} r)\} \rightarrow S$  as  $T \rightarrow 0$ .

Thus,  $Q = S(S+1)$  at  $T = 0$ .

## APPENDIX 2

### Q(T=0) FOR CLASSICAL VECTOR SPINS

It was shown by Mookerjee [392] that in the spin glass phase

$$Q_{xx} = Q_{yy} = Q_{zz} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dh_x dh_y dh_z \left(\frac{h_z}{h}\right)^2 P(h) \left[\coth(\beta h) - \frac{1}{\beta h}\right]$$

At  $T = 0$ ,  $Q_{xx}(T=0) = Q_{yy}(T=0) = Q_{zz}(T=0) = Q(T=0)$

$$\begin{aligned} &= \frac{1}{(2\pi cJ_1^2 Q)^{3/2}} \iiint dh_x dh_y dh_z \left(\frac{h_z}{h}\right)^2 e^{-h^2/2cJ_1^2 Q} \\ &= \frac{2\pi}{(2\pi cJ_1^2 Q)^{3/2}} \int_0^{\infty} dh \int_0^{\pi} d\theta h^2 \sin\theta \cos^2\theta e^{-h^2/2cJ_1^2 Q} \\ &= \frac{2}{3} \cdot \frac{1}{(2\pi)^{1/2} (cJ_1^2 Q)^{3/2}} \int_0^{\infty} h^2 e^{-h^2/2cJ_1^2 Q} dh \end{aligned}$$

Putting  $\frac{h^2}{2cJ_1^2 Q} = x,$

$$\begin{aligned} Q_{xx}(T=0) &= Q_{yy}(T=0) = Q_{zz}(T=0) = \frac{2^{1/2}}{3(2\pi)^{1/2}} \int_0^{\infty} \sqrt{x} e^{-x} dx \\ &= \frac{1}{\sqrt{\pi}} \cdot \frac{2}{3} \cdot \Gamma(3/2) = \frac{1}{3} \cdot \text{for } S^2 = 1 \end{aligned}$$

In the general case,

$$Q_{xx} = Q_{yy} = Q_{zz} = \frac{S^2}{3} \text{ at } T = 0.$$